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Generation IV Roadmap

Crosscutting Fuels and Materials R&D Scope Report

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Crosscutting Fuels and Materials R&D Scope Report

1. INTRODUCTION

The Generation IV Roadmap technical working groups (TWGs) have completed Research and Development (R&D) Scope Reports that identify the R&D necessary to prepare the selected Generation IV concepts for commercialization. For each concept, the R&D Scope Reports identify R&D gaps and recommend R&D tasks to address those gaps. The gaps are classified as either necessary for establishing concept viability or necessary for achieving desired performance. Relative priority or urgency is also indicated.

Before the execution of the Roadmap process, the Roadmap Integration Team and the Generation IV Roadmap NERAC Subcommittee anticipated that many of the R&D tasks required for different concepts would have similarities of some nature. The similarities are such that multiple concepts would benefit from a certain set of R&D tasks or that the R&D tasks would require similar techniques or experimental facilities. Consolidating such R&D into a single program is intended to reduce the required resources necessary to complete the combined set of objectives and to allow a broader look at technical issues that are similar. The Roadmap participants confirmed this during the evaluations leading to concept selection and during the preparation of the R&D Scope Reports. This report comprises the input of the Generation IV Fuel and Materials Crosscut Group (FMCG) to the Generation IV Roadmap Report and provides the Group recommendations for "crosscutting" R&D activities (or activities that are similar in nature).

Section 2 of this report provides a brief synopsis of R&D tasks identified in the TWG R&D Scope Reports and is intended to provide the reader with a sense of relevant R&D activities described in those reports. Section 3 discusses the opportunities for crosscutting R&D, and Section 4 then describes specific tasks. These R&D tasks are proposed in lieu of separate tasks described in the R&D Scope Reports to combine separate tasks or to supplement such tasks.

Another source that is used to identify crosscutting R&D opportunities is the documentation of the Higher Temperature Reactor Materials Workshop, which was sponsored in 2001 by the U.S. Department of Energy (DOE) Office of Nuclear Energy, Science, and Technology and Office of Basic Energy Sciences. The results are summarized in a report that is provided as Appendix A. Section 4 includes R&D tasks suggested by the Higher Temperature Reactor Materials Workshop, in addition to crosscutting R&D tasks identified by the FMCG.

2. SYNOPSIS OF FUELS AND MATERIALS R&D RECOMMENDED IN THE R&D SCOPE REPORTS

The R&D gaps for each of the selected Generation IV concepts and R&D proposed to address those gaps are described in the TWG R&D Scope Reports. This section provides a synopsis of those tasks that are related to fuels and materials technology. The intent of this section is not to describe these tasks in detail, but rather to identify the tasks that warrant a crosscutting R&D approach. The following synopsis is organized by concept.

2.1 Supercritical Water Reactor

Fuels and materials research for the Supercritical Water Reactor (SCWR) concept is concentrated in the areas of corrosion and stress corrosion cracking, radiolysis and water chemistry, dimensional and microstructural stability and strength, and embrittlement and creep resistance of fuel cladding and structural materials. Candidate alloys include austenitic iron (3xx series stainless steels) and nickel-base (solid solution: alloys 600, 690, 800, and precipitation-strengthened: 625, 718) alloys, ferritic-martensitic alloys (HT-9, T-91), and oxide dispersion strengthened alloys of either ferritic or austenitic structure. Critical R&D needs will focus on the temperature range of 280–620°C and irradiation damage dose ranges of 10–30 displacements per atom (dpa) (thermal spectrum) and 100–150 dpa (fast spectrum) and are categorized as follows.

The Supercritical-Water-Cooled Reactor (SCWR) system features two fuel cycle options: the first is an open cycle with a thermal neutron spectrum reactor; the second is a closed cycle with a fast-neutron spectrum reactor and full actinide recycle. Both options use a high-temperature, high-pressure, water-cooled reactor that operates above the thermodynamic critical point of water (22.1 MPa, 374ºC) to achieve a thermal efficiency approaching 44%. The fuel cycle for the thermal option is a once-through uranium cycle. The fast-spectrum option uses central fuel cycle facilities based on advanced aqueous processing for actinide recycle. Viability of the fast-spectrum option depends upon successful identification or development of materials for that application. In either option, the reference plant has a 1700-MWe power level, an operating pressure of 25 MPa, and a reactor outlet temperature of 550ºC. Passive safety features similar to those of the simplified boiling water reactor are incorporated. Note that the balance-of-plant is considerably simplified because the coolant does not change phase in the reactor.

2.1.1 Corrosion and Stress Corrosion Cracking

- Corrosion and stress corrosion cracking as a function of temperature, dissolved gases, and water chemistry
- Effects of irradiation on corrosion and stress corrosion cracking
- Composition and structure of corrosion films.

2.1.2 Radiolysis and Water Chemistry

- Radiolysis mechanism in supercritical water over a range of temperatures
- Chemical potentials and recombination rates of H2 and O2 and various radicals over a range of temperatures

- Effects of radiation on radiolysis yields and formation and reaction of other species by radiolytic processes.

2.1.3 Dimensional and Microstructural Stability

- Void nucleation and growth and the effect of He on voids and bubbles
- Evolution of dislocation and precipitate microstructure and radiation-induced segregation (RIS)
- Irradiation growth and stress relaxation.

2.1.4 Strength, Embrittlement, and Creep Resistance

- Tensile properties and time-dependent plasticity as a function of dose and temperature
- Creep, creep rupture, and creep-fatigue interaction
- Facture toughness, ductile-to-brittle-transition temperature and He embrittlement
- Microstructure-mechanical properties interaction under design-basis accident conditions.

2.2 Gas-Cooled Fast Reactor

The gas-cooled fast reactor (GFR) concept is proposed to combine the advantages of high-temperature gas-cooled reactors (such as efficient direct conversion with a gas turbine and the potential for application of high-temperature process heat) with the sustainability advantages that are possible with a fast-spectrum reactor. The latter includes the ability to fission all transuranics and the potential for breeding. The GFR is part of a consistent set of gas-cooled reactors that includes a medium-term Pebble Bed Modular Reactor-like concept, or concepts based on the Gas Turbine Modular Helium Reactor, and specialized concepts such as the Very High Temperature Reactor, or actinide burning concepts. The GFR concept is proposed to employ a direct helium Brayton cycle, utilizing core outlet temperatures of about 850°C. The GFR concept employs technology from the Pebble Bed Modular Reactor or Gas Turbine Modular Helium Reactor concepts where possible, but will require additional developments in fuel and materials to be successful.

Design parameters for the GFR that are relevant to fuels and materials include the following:

Average power density (but 100 MW/m^3 is a next objective)	55 MW/m^3
Core volume fractions (fuel, coolant, structure)	50%/40%/10%
Burnup target (but 15% FIMA is a next objective).	5% FIMA

In order to achieve the necessary high-power density and an ability to retain fission gases at high burnup and at high temperature, the fuel proposed for the GFR is a composite ceramic (CERCER) with closely packed and coated actinide carbide kernels or fibers. Alternative fuel concepts include fuel particles with large kernels and thin coatings and ceramic-clad solid solutions. Nitride compounds, enriched 99.9% in N-15, are considered as an alternative to the actinide carbide for the fuel kernel. In summary, Technical Working Group 2 proposed the following list of fuel materials for consideration:

2.2.1 Actinide Fuel Kernels

- (U,Pu)C
- (U,Pu)N

2.2.2 Inert Matrix Materials

- SiC-alpha, beta; ZrC; TiC
- ZrN, TiN, AlN, Si_3Ni_4
- MgO. $ZrYO_2$, CaO, Y_2O_3
- Cr, Zr, V, intermetallics.

2.2.3 Proposed R&D Tasks for GFR Fuels

- 2002–2004: acquisition of basic data on inert materials and actinide compounds; definition of reference and backup concepts.
- 2005–2012: irradiation testing in existing reactors.
- 2012–2020: irradiation testing of prototype fuel subassemblies in GFR-representative conditions.

2.2.4 Proposed Fuel Fabrication Development Tasks

- Fuel fabrication using innovative techniques is proposed, including vapor deposition and impregnation.
- Development of processes for fabricating actinide kernels or fibers
- Development of techniques for depositing actinide compounds and inert materials and for characterizing such deposits
- Development of robust techniques for fabricating CERCER and CERMET fuels as part of a recycle process
- Development of ceramic claddings or thick coatings to provide a barrier against fission product release.

2.2.5 Considerations for GFR Materials R&D

High-temperature materials are a recognized need for GFR application. Technical Working Group 2 has proposed basic research studies to complement an analysis of candidate materials for GFR application. In-core and out-of-core components located in the vessel must be capable of withstanding high, fast neutron exposures and temperatures that can reach as high as 1,600°C in some accident scenarios. Ceramic materials are the proposed reference for in-core use, with composite CERMETs or intermetallic compounds suggested as alternatives. High-temperature ceramic-based cladding is proposed for pin-type fuel options. Metal alloys are proposed as reference for out-of-core use.

The primary applications to be addressed in the GFR materials R&D program are:

- Fuel particle coatings and particle-containing baskets

- Inert matrix, fuel casings, and gas tubes for the composite fuel concept
- Cladding materials for the solid fuel concept
- Internal structures including upper and lower structures, shielding, the core barrel, grid plate, the gas duct shell, and the hot gas duct
- Pressure vessels and cross vessel.

Proposed materials for in-core application include the following:

- Preferred: SiC, ZrC, TiC, NbC
- Alternatives: ZrN, TiN, MgO. $ZrYO_2$ Cr, Zr, V, intermetallics such as Zr_3Si_2.

Proposed materials for out-of-core application include the following:

- Internal structures: ferritic-martensitic steels, coated or uncoated austenitics, other Fe-Ni-Cr bases (such as Inco 800), Ni-base alloys
- Pressure vessels: 2 1/4 Cr steels and 9-12 Cr ferritic-martensitic steels.

Selection of specific materials for in-core and out-of-core applications will be based on the best compromise of the following attributes:

- Fabricability and joining capability
- Physical, neutronic, thermal, and mechanical properties; initial characteristics and assessment of degradation of properties under neutron flux and dose (high for in-core, materials, low-to-moderate for out-of-core materials)
- Microstructure and phase stability under irradiation
- Irradiation creep and swelling properties
- Out-of-pile and in-pile compatibility with helium coolant (including impurities) and actinide compounds (for inert matrix materials).

2.3 Very High Temperature Reactor

The very high temperature reactor (VHTR) is a next step in the evolutionary development of high-temperature gas-cooled reactors. The key feature of this concept is the specification for an outlet temperature of 950 to 1,100°C. Review of the concept description and the R&D proposed by Technical Working Group 2 clearly indicates that fuels and materials selection for high-temperature service will be a key design issue for this concept. However, materials suitable for these applications, particularly for in-core applications, cannot be specified with certainty.

Core outlet temperatures less than 1,000°C can likely be accommodated with metallic materials and SiC-based TRISO fuels. However, outlet temperatures above 1,000°C will require different materials for the reactor pressure vessel, for core internals, for cooling pipes and heat exchangers, and for fuels.

The TRISO particle is proposed as the fuel design for the VHTR. However, requirements call for reliable operation at conditions considerably more severe than previous TRISO application. Specific requirements for fuel include the following:

- Increase of coolant outlet temperatures from 850°C up to 950–1,100°C (which results in an increase in fuel temperature)
- Increase of tolerable accident temperature limits beyond 1,600°C, up to 1,800°C
- Increase in maximum burnup from 80 GWd/MTU to 150–200 GWd/MTU.

2.3.1 Proposed R&D Tasks for VHTR Fuels

Application of ZrC coatings, which appear to better mitigate the diffusive release of fission products at high temperature than do SiC coatings, is proposed for VHTR fuel. It has also been suggested that the fuel might be redesigned to better accommodate increased amount of fission gas pressure. Furthermore, the incorporation of a burnable poison might be required to reduce burnup reactivity swing associated with high-burnup operation.

Specific R&D activities include the following:

- Development and evaluation of high-temperature coatings such as ZrC
- Development and irradiation testing of high-burnup fuel designs
- Accommodation of a burnable absorber into a fuel design.

2.3.2 Proposed R&D Tasks for VHTR Materials

The VHTR systems will operate at higher temperatures than other gas-cooled reactors and will require different materials for important components. For example, reactor pressure vessels will operate at temperatures greater than 450°C, whereas light water reactor pressure vessels operate at about 300°C and the High Temperature Test Reactor in Japan operates at about 400°C. Therefore, new metallic materials will likely be required for VHTR pressure vessels. Graphite can be used for core internals, as for previously operated gas-cooled reactors, but further improvements in graphite properties, such as oxidation resistance and structural strength, will be necessary. Other core internals and cooling system components—such as the intermediate heat exchanger, hot gas ducts, and isolation valve sheets—will require ceramic materials. Carbon-carbon composites are being considered for control sheaths for VHTR concepts that employ prismatic graphite blocks for core structures. Other promising ceramics that might be considered include fiber-reinforced ceramics, sintered aloha SiC, oxide composite ceramics, and compound materials.

The R&D activities proposed for VHTR materials development include:

- Acquisition of mechanical and thermal properties data, fracture behavior information, and oxidation data
- Irradiation tests
- Post-irradiation heat-up test of fuel to evaluate accident-related transient behavior

- Development of models of materials behavior and a stress analysis code that considers anisotropy.

2.4 Sodium-Cooled Reactor

The sodium-cooled reactor concept proposed for Generation IV is based on familiar sodium-cooled reactor technology that was developed worldwide until the mid-1990s and remains under development in Japan and, to some extent, France. The reactor will be fueled with either U-Pu-Zr alloy fuel supported by pyrometallurgical reprocessing or $(U,Pu)O_2$ fuel supported by advanced aqueous reprocessing. Much of the fuels and materials-related R&D for those concepts is considered to be performance related, but some is considered important for system viability.

The concepts proposed for Generation IV call for incorporation of recycled actinides into the recycled fuel to allow actinide consumption. Therefore, the applicability of the U-Pu-Zr and $(U,Pu)O_2$ performance databases to U-Pu-Zr-MA and $(U,Pu)O_2$-MA_xO_y must be determined through irradiation and safety testing of minor-actinide-bearing fuel. In particular, the effect of minor actinide constituents in metal alloy and residual fission products fabricated into recycled fuel on fuel-cladding interdiffusion must be assessed.

HT9 is an adequate cladding and duct material for the U-Pu-Zr-fueled sodium-cooled reactor with low-to-moderate core outlet temperature; however, selection of a material with improved high-temperature strength and creep resistance would improve performance and further increase the safety margin. The oxide-fueled sodium-cooled reactor is proposed with an outlet temperature (550°C) that is higher than previous experience with sodium-cooled reactors. Therefore, an alloy with better high-temperature properties will be necessary if the oxide-core concept is to perform as proposed. Oxide dispersion-strengthened ferritic alloys have been proposed for this application.

The presence of minor actinides (and residual fission products for the case of the U-Pu-Zr-MA alloy) in recycled fuel feedstock will require remote, shielded fabrication processes. Furthermore, the retention of volatile americium is an issue for the fabrication of metal fuel, because the familiar injection casting process employs elevated temperature at which americium volatilizes from an alloy melt. Therefore, R&D on improved fabrication methods for oxide fuel and for metal alloy fuel is proposed. Specific R&D tasks proposed by Technical Working Group 3 include development of the Simplified Pelletizing Method as a means to facilitate the remote application of an oxide pellet fabrication technique and development of the vibrational compaction (vi-pac) technique for fabricating oxide fuel.

The reduction or elimination of actinide losses to secondary waste streams resulting from fabrication will be essential if the concept is to meet Generation IV goals for sustainability. This implies that, for metal alloy fuel, interactions between fuel and recycle or fabrication components must be minimized and processes that release considerable amounts of contamination must be modified. Therefore, Technical Working Group 3 has proposed development of improved crucible materials and re-usable injection casting molds. Reduction of losses from oxide fuel fabrication would be achieved through development of alternative techniques such as the Simplified Pelletizing Method or the vi-pac technique.

Fuels research for the oxide-fueled sodium-cooled reactor with advanced aqueous reprocessing is motivated mainly by the newly envisioned missions, namely recycling and consuming of minor actinides. Critical R&D needs are as follows.

2.4.1 Irradiation Performance

- Irradiation testing of selected minor actinide-bearing mixed oxide (MOX) fuel compositions

- Oxide-dispersion strengthened cladding alloy development
- Increasing fuel burnup.

2.4.2 Modeling and Code Development

- Evaluate irradiation and safety performance of minor actinide-bearing fuel and update existing models
- Develop models for behavior of advanced cladding materials and incorporate into fuel performance codes.

Fuels research for metal-fueled sodium-cooled reactor with pyroprocessing focuses on the characterization of recycled fuel. Critical R&D needs are as follows.

2.4.3 Property Determination

The impacts of minor actinide additions on properties must be assessed.

- Confirmation of thermal conductivity, heat capacity, and thermal expansion of recycled fuel
- Investigation of fuel/cladding interdiffusion behavior with enhanced quantities of rare earth products and minor actinides.

2.4.4 Irradiation Performance

- Qualification irradiations of U-Pu-Zr, including 2-sigma conditions, to ensure that U-Pu-Zr behavior is bounded by a safety analysis for whole-core operation
- Evaluation of the effect of enhanced minor actinide and recycling impurity contents on irradiation performance lifetime
- Transient testing of high-burnup U-Pu-Zr, with and without minor actinide and reprocessing impurity additions, under specific transient overpower and loss-of-flow condition
- Development of cladding alloys with improved high-temperature strength over HT-9, but with similar resistance to swelling
- Development of cladding liners that reduce effects of fuel-cladding interdiffusion or improved cladding alloys that incorporate this feature in addition to high-temperature strength and swelling resistance.

2.4.5 Modeling and Code Development

- Development of a mechanistic, predictive model for fuel-cladding chemical interaction
- Development of a mechanistic, predictive model for fuel migration.

2.4.6 Out-of-Core Materials

Little additional research and development remain for the materials applied to the out-of-core structures of sodium-cooled reactors. The outstanding research and development are the development and/or selection of structural material for components and piping. The 12-Cr ferritic steels, rather than austenitic steels, are viewed as promising structural materials for future plant components. Critical R&D needs in terms of 12-Cr ferritic steels are as follows:

- Accumulation of material strength database focusing on the creep-fatigue
- Improvement of toughness and ductility
- Welding procedure development
- Elevated temperature strength data of welded joints
- Manufacturing technology development for thick plate and thin-walled heat transfer tube.

2.5 Lead Alloy Reactor Concepts

Several Pb or Pb-Bi reactor concepts have been proposed for Generation IV. The key features of these concepts include the application of Pb or Pb-Bi coolant to provide good heat transfer, natural convection characteristics, and increased coolant boiling temperature. Such characteristics make innovative approaches possible. For example, the Pb-Bi "battery" concept is proposed to provide a small modular reactor that can be implemented in countries requiring smaller increments of power and that do not wish to develop an indigenous fuel cycle capability. The core design is intended to allow long service between refueling, which would be facilitated with a modular, cartridge-type core/fuel assembly. The initial designs would employ a core outlet temperature of about 550°C or lower, but the proposal calls for eventual development of the concept to take advantage of the thermophysical properties of Pb or Pb-Bi coolant by employing a 750 to 800°C outlet temperature.

A key challenge to the development of lead alloy concepts is the selection of structural materials, including cladding and in-core materials that are compatible with Pb or Pb-Bi at elevated temperatures. In fact, establishing the technology base for working with such coolants is considered a necessary first step in developing the class of concepts. Russian technologists have identified some ferritic-martensitic alloys as appropriate for in-core use with peak temperatures around 600°C and coolant velocity under 2 m/s, and some austenitic alloys for ex-core use. The suitability of such alloys for Pb/Bi battery application must be verified. Further work will be required to select in-core structural materials that allow higher outlet temperature operation. Some ceramic materials (e.g., SiC) and refractory metals have been proposed for this application, but little is known about such use of these materials. In addition, use of high outlet temperatures for alternative energy conversion technologies will likely require selection of additional high-temperature materials for use in heat exchangers or for energy conversion components.

Nitride fuel, (U,Pu,)N or (U,Pu,MA)N, is proposed for the high-outlet-temperature variant and for most of the 550°C outlet-temperature concept variants. Although (U,Pu)N was considered previously as an advanced fuel for liquid metal breeder reactor use, the fuel form remains relatively immature. Nitride pellet fabrication is possible using conventional techniques; however, recycle of minor actinides or accommodation of residual fission products from recycle might complicate the implementation of those techniques. Furthermore, the expected necessity for remote, shielded fabrication will motivate use of alternative processes, such as the Simplified Pelletizing Method or the vi-pac technique discussed for fabrication of oxide fuel for the sodium-cooled reactor concept. Another complication to the use of nitride fuel is the need to use nitrogen enriched in N^{15} in order to avoid substantial generation of C^{14}, which is a product of neutron capture in N^{14}.

The irradiation performance of nitride fuel must be established through irradiation testing in a prototypic environment (i.e., neutron spectrum, temperature, and coolant chemistry). Irradiation performance characteristics to be considered include fuel, fission product, and cladding interdiffusion at high temperature and high burnup; fuel fragmentation behavior; cladding performance in Pb or Pb-Bi; and post-breach behavior. The safety-related performance of nitride fuel must be evaluated through transient testing in overpower and undercooling conditions. Any reduction in safety margin due to fuel properties or behavior will be addressed through design.

2.6 Molten Salt Reactor

The Molten Salt Reactor (MSR) is unique among selected Generation IV concepts in that it uses a fluid fuel rather than a sold fuel. The fluid fuel is pumped through the core region of the reactor, where fuel volume, moderation, and reflection allow a critical reaction, and then it flows out of the core through a heat exchanger and systems that remove fission products for the fuel. The liquid fuel is a low-melting-temperature mixture of salts, which includes uranium and may include thorium or transuranics. Because the fuel is liquid, the issues associated with fuel performance are considerably different than those for solid fuels. However, the higher outlet temperatures of the system imply similar materials property concerns as other Generation IV systems, with the additional challenge of molten salt compatibility.

The materials proposed for MSR application are metal alloys and graphite. The metal alloys include INOR 8, Hastelloy B and Hastelloy N, Ni-based alloys, and niobium-titanium alloys. Of these, a modified Hastelloy N alloy is preferred. An addition of about 2 wt.% titanium appears to reduce irradiation embrittlement due to helium bubble formation and increase resistance to intergranular tellurium attack. Graphite is proposed as an in-core structural material, to define the salt flow pattern, and as a moderator. The challenges for graphite use in this application are radiation damage that leads to dimensional changes in the graphite, salt penetration into the graphite, and xenon absorption into the graphite, which can increase fission product poisoning effects and reduce the breeding/conversion ratio.

The R&D plan prepared by Technical Working Group 4 proposes a viability research phase that includes materials compatibility testing in a controlled chemistry test loop. These tests would assess the ability of the materials to resist chemical attack and corrosion by the fuel at high temperature. Insertion of a similar test loop into a thermal-spectrum test reactor would allow additional assessment of radiation damage effects. However, the ability to test with representative fuel compositions and simulated fission products must be further assessed.

The proposed Performance R&D phase calls for additional testing of candidate structural materials in molten fuel salt test loops. In particular, such testing with materials such as Hastelloy N would include in-pile testing in loops. The tests would allow assessment of thermal gradient-enhanced corrosion effects in molten salt, including measurements of dissolution parameters, diffusion coefficients, kinetic coefficients toward temperature, and redox potential. Development of redox control techniques would be particularly important for materials compatibility in the secondary loop. The extent of tellurium embrittlement and the effect of radiation damage on mechanical properties would also be assessed.

Further development of graphite is deferred until the Performance R&D phase. The development of sealing techniques to stop xenon penetration will be undertaken. Use of high-quality, fine-grained graphite is specified to reduce salt penetration. However, dimensional changes induced by radiation damage are an important consideration, as those dimensional changes require somewhat frequent replacement of in-core graphite structure (e.g., every 4 years for the Molten Salt Breeder Reactor).

Application of fabrication techniques to produce desired graphite microstructures is suggested as a means to reduce radiation effects, and research and development into the efficacy of such techniques are proposed.

2.7 Summary of Requirements and Candidate Materials

The following general observations are made regarding fuels and materials for Generation IV systems:

- Even among concepts with different coolants, many application requirements have important similarities (such as temperature, stress, or neutron spectrum), prompting selection of similar materials, or classes of materials, for various concepts. This is underscored by Table 1, which indicates classes of materials considered as candidates for specific concepts.

Table 1. Summary of classes of materials considered for specific Generation IV applications.

	Fuel Materials						Structural Materials						
System	Oxide	Nitride	Metal	Carbide	TRISO Particle	Fluoride (liquid)	Ferritic-martensitic Stnlss Steel Alloys	Austenitic Stnlss Steel Alloys	Oxide Dispersion Strengthened	Ni-based Alloys	Graphite	Refractory Alloys	Ceramics
GFR	–	–	–	[a]P	–	–	P	P	P	P	–	P	P
Pb Alloy	–	P	–	–	–	–	P	P	[b]S	–	–	S	S
MSR	–	–	–	–	–	P	–	–	–	P	P	S	S
Na-LMR	P	–	P	–	–	–	P	P	P	–	–	–	–
SCWR-Thermal	P	–	–	–	–	–	P	P	S	S	–	–	–
SCWR-Fast	P	–	S	–	–	–	P	P	S	S	–	–	–
VHTR	–	–	–	–	P	–	S	–	–	P	P	S	P

a. P = Primary option.

b. S = Secondary option.

- All systems project in-service and off-normal temperatures that are beyond current nuclear industry experience, as well as most previous experience with developmental systems.

- Most systems call for use of fast and epithermal neutron spectra (to accommodate sustainability objectives through higher conversion ratios and the ability to consume transuranic elements), which will challenge materials performance with increased radiation damage.

- All systems require relatively long service lifetimes of components and relatively high burnup capability of fuels—in environments with increased temperature and radiation damage.

- Many of the system concepts are proposed with an acknowledgement that suitable materials of construction for various components are not yet identified or are proposed based on notional impressions of material performance. A simple selection and qualification of materials is unlikely and a substantial materials-related R&D program will be necessary for concept viability.

- The lack of data for proposed materials relevant to the challenging conditions suggests that a broad-based materials R&D program will serve the development of multiple concepts.

Fuels and materials that meet the requirements of Generation IV systems must be identified, and databases sufficient to support design and licensing of Generation IV systems must be established. A summary of the fuels and materials options proposed for each of the concept sets is provided in Table 2. For applications where the responsible TWG did not propose a material of construction, the FMCG has suggested a material or class of materials for initial consideration. The table reflects initial suggestions, and there are little or no data to support the recommendation of a specific alloy or material. It should be noted that some classes of materials, such as ferritic-martensitic oxide dispersion-strengthened alloys (ODSs), ferritic-martensitic stainless steels (F-M), ceramic materials, or refractory alloys, are proposed for multiple applications in multiple concepts.

Table 2. Fuels and materials proposed for Generation IV systems.

				Structural Materials	
System	Neutron Spectrum, T_{outlet}	Fuel	Cladding	In-core	Out-of-core
GFR	Fast, 850°C	MC/SiC	Ceramic	Refractory metals and alloys, ceramics, ODS Vessel: F-M	Primary Circuit: Ni-based superalloys 32Ni-25Cr-20Fe-12.5W-0.05C Ni-23Cr-18W-0.2C F-M w/therm. barriers Turbine: Ni-based alloys or ODS
Pb Alloy	Fast, 550°C and Fast, 800°C	MN	High-Si F-M, ceramics, or refractory alloys		High-Si Austenitics. Ceramics or refractory alloys
MSR	Thermal, 700°C	Salt	Not applicable	Ceramics, refractory metals, High-Mo Ni-base alloys (e.g., INOR-8), Graphite, Hastelloy N	High-Mo Ni-base alloys (e.g., INOR-8)
Na-LMR (Metal)	Fast, 520°C	U-Pu-Zr	F-M (HT9 or ODS)	F-M ducts 316SS grid plate	Ferritics, austenitics
Na-LMR (MOX)	Fast, 550°C	MOX	ODS	F-M ducts 316SS grid plate	Ferritics, austenitics
SCWR-Thermal	Thermal, 650°C	UO_2	F-M (12Cr, 9Cr, etc.) (Fe-35Ni-25Cr-0.3Ti) Incoloy 800, ODS Inconel 690, 625, and 718	Same as cladding options	F-M
SCWR-Fast	Fast, 650°C	MOX, Dispersion	F-M (12Cr, 9Cr, etc.) (Fe-35Ni-25Cr-0.3Ti) Incoloy 800, ODS Inconel 690 and 625	Same as cladding options	F-M
VHTR	Thermal, 1,000°C	TRISO UOC in graphite compacts; ZrC coating	ZrC coating and surrounding graphite	Graphites PyC, SiC, ZrC Vessel: F-M	Primary Circuit: Ni-based superalloys 32Ni-25Cr-20Fe-12.5W-0.05C Ni-23Cr-18W-0.2C

F-M w/therm. barriers
Turbine: Ni-based alloys
or ODS

Abbreviations
F-M = Ferritic-martensitic stainless steels (typically 9 to 12 wt.% Cr)
ODS = Oxide dispersion-strengthened steel (typically ferritic-martensitic)
MN = (U,Pu)N
MC = (U,Pu)C
MOX = $(U,Pu)O_2$

3. OPPORTUNITIES FOR CROSSCUTTING R&D

A review of the R&D in the preceding synopsis reveals a number of opportunities for advantageous crosscutting R&D tasks or R&D approaches. This section describes those opportunities, which appear in categories that are defined for convenience or based on an important similar aspect.

3.1 General Observations Regarding Fuels and Materials R&D

Where possible, the Technical Working Groups have proposed a material or class of materials for each challenging application or component, based on experience with similar, less challenging nuclear applications or based on non-nuclear applications of similar technology. However, success of these materials for Generation IV application is uncertain. Furthermore, the materials property data and behavior data required for design and licensing are not available. The recommended approach is to initially consider a material, or class of materials, for each application based on established experience. If these initial material selections prove successful, then much of the Generation IV materials effort will simply be collection of properties data and assessment of behavior. However, if the initial selections do not meet requirements, then modifications to the material compositions or fabrication processes will be attempted. Ultimately, development of new alloys or materials may be required. Materials development and characterization programs in the past have proven costly and lengthy. However, the opportunity for international collaboration and for addressing the needs of concepts through crosscutting R&D tasks should render the effort less formidable.

Consideration of the full range of material needs for the Generation IV concepts indicates that many will benefit from a single body of R&D activities. Although service applications for analogous components will have differences that are specific to the concept, many aspects of the service environments are similar across sets of concepts. Of these aspects, service temperature and neutron spectrum will have the greatest influence on material performance and component lifetime. In fact, experience with materials performance has clearly indicated that irradiation properties are as important to component lifetime as other environmental degradation phenomena, such as corrosion in light water reactors. Today, Generation IV developers have the advantage of a considerable and relevant technology base for non-nuclear applications, and the benefit of an enhanced understanding of mechanisms that control irradiation behavior and high temperature performance of materials, that was not available to the previous development programs. Therefore, the Generation IV Program offers a unique opportunity for addressing the development needs of several concepts with a set of experiments designed to determine properties and irradiation behavior at common but important service conditions.

3.2 Structural Materials Properties and Behavior

Evaluating basic physical and mechanical properties of several candidate in-core and ex-core materials constitutes a crosscutting opportunity that addresses several of the concepts. According to

Table 2, the coolant outlet temperatures of these six concepts exceed that of current by at least 190°C, and in some cases, by over 600°C. Similarly, the fast spectrum concepts will result in radiation damage doses to core internals that exceed those in current light water reactors by as much as a factor of 10. The combination of higher operating temperature and higher radiation damage dose places these concepts into a parameter space for which little or no commercial plant operating data are available.

Over the temperature range 300–600°C, candidate materials will require sufficient strength and resistance to diffusion-driven processes such as radiation-induced segregation and precipitation, void formation and growth, dislocation loop growth, creep, fatigue and high temperature corrosion, and stress-induced corrosion cracking processes. In this temperature regime, all microstructure features change quickly with temperature. Little is understood about the complex interactions that could occur between microstructure features such as dislocations, voids/bubbles, RIS, and precipitates when each is a very sensitive function of temperature. The interplay between these features and their relative sensitivities to temperature will be important to understand for this alloy system to be applied in this temperature regime. Candidate alloys in this temperature range include austenitic iron- and nickel-base alloys, ferritic-martensitic (F-M) alloys, and OD alloys.

Above 600ºC, thermal mechanisms, rather than irradiation–induced mechanisms, dominate the behavior of most metallic alloy systems of interest. For example, the dominant creep mechanism becomes thermal, rather than irradiation creep. In fact, in alloys for which the operative temperature range exceeds a homologous temperature (T/T_m) of 0.6, the radiation damage is increasingly annealed away. Conventional austenitic, ferritic, or F-M steels cannot be used above 600°C with any significant level of applied stress. The ODS F-M steels exhibit promising improvements in creep resistance up to ~800°C, but remain the subject of ongoing research. The primary alternatives are materials such as precipitate-strengthened iron- or nickel-base superalloys or refractory alloys of molybdenum, niobium, and tantalum, which are proposed because of their high strength and good high-temperature creep resistance. Unfortunately, little is known about the microstructural development of either alloy class under irradiation at these temperatures. Finally, inherent toughness problems, limited understanding of radiation-induced material changes, and a high sensitivity to oxygen-impurity pickup that can lead to further embrittlement challenge the practical use of refractory alloys.

Materials issues for applications at temperatures up to 900°C also apply at temperatures exceeding 900°C, with an increasing level of severity. Possible alternate materials may have acceptable high-temperature performance, but there are insufficient data to design complex engineering structures, particularly those that will be exposed to neutron radiation. Of the potential metal-based systems, only tungsten- and molybdenum-based systems are believed to have the potential to operate in this temperature range. Little or no data exist on the development of microstructure under irradiation for either of these systems. There is virtually nothing known about void formation, swelling, RIS, dislocation microstructure or precipitate formation, and stability under irradiation at temperatures above 900°C. The extreme temperatures also present problems for conducting controlled experiments in existing irradiation facilities.

The materials to be selected for balance-of-plant components will not experience neutron radiation fields, as will the in-core components. However, operating temperatures that are higher than those to which current balance-of-plant components (e.g., heat exchangers) are subjected, and the additional compatibility issues that are introduced with alternative energy products, will challenge such ex-core materials. For example, generation of hydrogen or hydrogen-bearing fuel products will entail relatively high operating temperatures as well as environments that are potentially corrosive or embrittling to some materials.

A broadly applicable R&D program is proposed to study and quantify the properties of materials proposed for Generation IV application. The information gained will be used to derive new materials necessary to meet requirements and to support system design and licensing. Specific research program objectives required in order to qualify the proposed classes of materials are as follows:

- Assessment of void nucleation and growth and the effect of He production on void stability and growth and He bubble nucleation and growth as a function of dose (up to ~30 dpa in a thermal spectrum and up to 150 dpa in a fast spectrum) as a function of temperature.

- Development of the dislocation microstructure, precipitate microstructure, and radiation-induced segregation as a function of dose (up to ~30 dpa in a thermal spectrum and up to 150 dpa in a fast spectrum) as a function of temperature. The stability of oxide particles in irradiated ODS alloys would be included in this task.

- Knowledge of growth or irradiation-induced distortion as a function of dose (up to ~30 dpa in a thermal spectrum and up to 150 dpa in a fast spectrum) as a function of temperature.

- Knowledge of irradiation-induced stress relaxation as a function of tension, stress, material, and dose.

- Tensile properties (yield strength, ultimate tensile strength, elongation, reduction in area) as a function of dose over the range 10–30 dpa (thermal design) and 100–150 dpa (fast design) as a function of temperature.

- Creep rates (primary and secondary) in candidate alloys in the dose range 10–30 dpa (thermal spectrum) and 100–150 dpa (fast spectrum) over a range of temperature and applied stress.

- Creep and creep rupture mechanisms for the same dose, temperature, and stress conditions as used for creep rate measurements.

- Creep-fatigue interactions and dependence on cyclic loading frequency, baseline versus load following, effects of RIS on creep-fatigue.

- Fatigue crack growth rate data in irradiated materials.

- Time-dependence of plasticity and high temperature plasticity.

- Microstructural impact of creep-fatigue and feed back loop.

- Helium embrittlement at operating temperatures (slow strain rate testing).

- Fracture toughness as a function of temperature and ductile-to-brittle-transition-temperature (DBTT) for each of the candidate alloys in the unirradiated condition (quasi-static and dynamic strain rates).

- Fracture toughness as a function of irradiation temperature and dose.

- DBTT and helium embrittlement as a function of dose and irradiation temperature.

- Interaction between radiation-induced aging and fracture toughness/DBTT.

- Changes in microstructure and mechanical properties following design basis accidents. Given the transient performance regimes, structural materials must be able to withstand accident conditions without compromising their mechanical property integrity (creep resistance, strength, embrittlement).

3.3 The Fuel Development Process

Based on previous experience with development of fast reactor fuels and based on the varying maturity of the proposed fuel forms, varying needs for fuel development are envisioned. In general, a long-term program to develop fuels for deployment entails the following activities: (1) fabrication process development, (2) property measurement and assessment, (3) irradiation testing and safety demonstration, and (4) modeling and predictive code development.

Four phases of development are proposed, which include the above activities to varying degrees. The phases of fuel development are as follows:

- Fuel candidate selection
- Concept definition and feasibility
- Design improvement and evaluation
- Fuel qualification and demonstration.
- The different fuels proposed for Generation IV systems have been developed through varying stages of the four-phase sequence. However, because many of the activities listed above are utilized across multiple phases, there will be similarities in necessary R&D for fuels with different maturity. The development for structural materials follows a similar path.

3.4 Fuel Materials Properties

Actinide recycle into a fast-spectrum reactor has been identified by the Fuel Cycle Crosscut Group as a means to manage the disposition of transuranic isotopes, which have long half-lives and dominate the calculated long-term dose associated with releases from repositories such as Yucca Mountain in the U.S. Therefore, the fast-spectrum Generation IV concepts will be developed to accommodate minor actinides in fuel or in special targets. The properties of minor actinide-bearing-fuel materials are not well established (although, for dilute minor actinide contents, the properties should not differ substantially from the base U-Pu-bearing materials). However, key properties—such as thermal conductivity, enthalpy, and melting temperatures—must be determined or verified. Other intrinsic characteristics, such as interdiffusion of fuel constituents under temperature gradients and interdiffusion with cladding constituents, must also be assessed. Because some fuel types are proposed for multiple concepts and because the sample preparation and measurement techniques are quite specialized, this set of activities presents a crosscutting R&D opportunity.

3.5 Irradiation Testing

The R&D plans for all concepts call for irradiation testing of fuels and materials for in-core components. A review of the spectrum and temperature conditions listed in Table 2 indicates that, independent of coolant type, many of the proposed irradiation environments and, therefore, testing environments would be similar. Furthermore, there are currently no operational test loops for testing with lead, helium, or supercritical water coolants in a fast-spectrum test reactor, nor a supercritical water test loop in a thermal-spectrum test reactor.

Many of the test reactors that had been used for irradiation testing in support of previous nuclear reactor R&D programs are no longer available. In fact, only a small number of fast-spectrum test reactors remain operational in the world—none in the United States. Furthermore, there are currently no operational test loops for testing with lead or helium coolants in a fast-spectrum test reactor. Although a larger number of thermal-spectrum test reactors remain available, the ability to fully test and qualify thermal-spectrum fuels is limited to a small number of reactors with larger irradiation test volumes. Although there are few gas-cooled test reactors (such as the High Temperature Test Reactor in Japan), the prospect for testing gas reactor fuel is improved by the ability to test such fuels in water-cooled test reactors using helium-cooled test vehicles; this has been accomplished, for example, at the Advanced Test Reactor and the High Flux Isotope Reactor in the United States.

The similar nature of the service conditions for sets of concepts and the limited availability of irradiation test facilities are two strong reasons to consider a well-coordinated Generation IV irradiation-testing program. This program would likely be comprised of tests and experiments at reactors in several countries, serving the requirements of several concepts. Post-irradiation examination and testing would be an important component of the program and would be the subject of specific interactions among participants. The applicability of post-irradiation testing for many structural components and joints envisioned for Generation IV in-core materials will enable the use of light ion irradiation for the purpose of conducting rapid, low-cost screening studies on a large number of alloys. Although the specific elements of the irradiation test program cannot yet be defined, a number of types of irradiations are envisioned, as described below.

Types of Irradiation Tests. The envisioned irradiation testing activities, many of which are described in the concept R&D sections of this report, are summarized below.

- Inert environment tests of unirradiated and pre-irradiated structural material samples at relevant temperatures to assess radiation effects on mechanical behavior (strength, creep, fracture toughness) over the temperature range of interest in the classes of materials proposed for all applications, independent of coolant-induced phenomena.

- Special-effects irradiation tests in laboratories simulating the effect of neutrons and fission products on material microstructures using ion beams. Such tests would be used as low-cost means for assessing microstructural evolution in structural materials or in matrix materials proposed for dispersion fuel concepts. These tests might include swift ion irradiation or fission product and helium implantation.

- Irradiation tests of material samples in prototypic neutron spectra and in flowing coolant loops (or flowing fuel loops, in the case of MSR R&D) are fundamental to assess the effects of environmental degradation (e.g., due to radiolysis-enhanced corrosion and in situ radiation damage) on materials properties and performance. Concept-specific corrosion and environment testing of pre-irradiated samples would provide a low-cost means of assessing the impact of radiation damage on environmental degradation of performance.

- Preliminary, low-cost tests of new fuel designs (either new fuel forms or new compositions) in a specially configured vehicle in a test reactor to identify irradiation performance issues.

- Irradiation tests of prototypically designed test fuels to determine fuel lifetime and life-limiting phenomena in proposed fuel designs.

- Irradiation tests of reference fuel designs at conditions of power and temperature that determine limits for safe and reliable operation of fuels. This information will be essential for supporting a licensing case for a first-of-a-kind reactor.

3.6 Transient Testing of Reactor Fuels

All Generation IV concepts will require transient testing, the nature of which will depend on the stage of development of a given fuel concept. Fuels that are in initial development stages will require transient testing, independent of design-basis accident issues, to understand transient response and to aid design changes that ensure required safety-related behavior. Fuel concepts that have matured to the point of reference designs will require transient testing under a range of design-basis accident conditions to determine mechanisms that lead to fuel failure and threshold conditions at which fuel failure occurs. Fuels with better-established performance databases will require testing at specific design-basis accident conditions to verify that behavior in the new Generation IV system is as expected, which will be an important step in qualifying the fuel for licensing.

These types of tests and experiments are typically performed in specialized test facilities—a transient testing reactor for overpower transients and certain undercooling conditions—and out-of-pile furnaces and test loops for other types of undercooling conditions. Many of the personnel and facilities that were previously employed in various countries for such testing are no longer available. This fact, and the similar nature of the test methods used for differing fuel types, suggests that a single transient test program, or a coordinated set of programs, will ensure a consistent approach to safety testing of the various fuel types and will provide an opportunity for efficient use of facilities and personnel. At this point, a specific test program cannot be proposed. However, the elements of the program can be envisioned to include overpower and undercooling tests of new fuel types in capsules containing prototypic coolants and design-basis accident tests in prototypic coolant, temperature, and power environments. For now, the only related cross-cutting R&D task proposed is the establishment of transient testing capability to serve the needs of all the Generation IV concepts. However, it is envisioned that developmental testing of similar fuel forms for multiple concepts can be accommodated as those needs are better clarified.

3.7 Fuel Fabrication Technique Development

As shown in Table 2, four of the proposed fuel types are ceramic pellet fuels. The R&D plans for the mixed oxide-fueled sodium-cooled reactor include development of the Simplified Pelletizing Method, which is intended to provide a fabrication scheme that is simpler and less contamination intensive than the currently used techniques. Because mixed oxide fuel is proposed as an option for the fast-spectrum Supercritical Water Reactor, the development of the technique will benefit that concept as well. Furthermore, the Roadmap participants believe that this technique can potentially be applied to the fabrication of other pellet type fuels—specifically, the mixed nitride fuel pellet options proposed for the lead-cooled reactor concept and the fast-spectrum variant of the Supercritical Water Reactor Concept. A small R&D task to consider whether and how the Simplified Pelletizing Method can be extended to nitride fuel fabrication is proposed in Section 4.

The vibrational compaction (vi-pac) method has been proposed as an alternative for fabrication of oxide fuel for the Na-cooled reactor. The technique was studied previously in several countries, including the U.S., France, and Russia. However, only Russia is currently employing the technique, which is used to fabricate fuel for the BOR-60 reactor. The technique is appealing for Generation IV application because it allows relatively simple fabrication of ceramic fuel in a shielded hot cell environment. However, the resulting fuel form is different than pellet fuel, in that the ceramic powders form a packed-bed fuel column within the cladding, and the packed bed fuel column behaves differently than pellet fuel. The technique might also be applicable to fabrication of nitride fuel, and the FMCG recommends that be considered. The R&D needs associated with the vi-pac technique include determining the particle size distribution and vibration conditions required for producing the desired packed fuel column. Preparation of the particles within the desired size distribution might be difficult in a shielded hot cell, so additional work to develop a suitable technique might be necessary. Finally, the sensitivity of fuel performance to deviations in fabrication parameters must be assessed with irradiation testing.

3.8 Process Materials Selection and Development

Although much of the emphasis of this report is on fuels and materials for reactor systems, it is noted that other associated technologies will present materials challenges as well. In particular, the success of the fuel recycle technologies will depend upon selection of construction materials that allow processing under harsh environmental conditions such as high temperature and high radiation fields. In addition to successful functional performance, the selected materials must also exhibit a low propensity for interaction with the recycled fuel media, which will be essential to minimizing the amount of actinides sent to secondary waste streams. Therefore, for recycle technology that is developed for potential application to multiple Generation IV concepts, an associated crosscutting materials R&D effort is proposed.

The requirements for new process materials have not been fully considered, but some applications are now evident. For example, injection casting of metal fuel will benefit from development of reusable molds that will reduce the amount of residual actinides lost to mold waste. Many recycle operations in the pyrometallurgical process will be improved with better crucible materials (e.g., crucibles for consolidation of cathode deposits or for induction melting prior to injection casting). Similarly, waste resulting from ceramic fuel fabrication processes can be reduced if more durable die materials are developed. The details of the proposed tasks are the same as those proposed in the concept-specific R&D plans. Here, the FMCG simply recommends that these efforts be applied to multiple concepts.

3.9 Materials Phenomena Modeling

The design of new alloys for Generation IV systems is a highly intensive exercise requiring considerable expenditure of resources on sophisticated experimental systems. More experimental programs will be demanded than can be supported, thus limiting the amount of data or perhaps the degree to which prototypic conditions and geometries can be studied. The capability to model material properties and performance will therefore be highly valuable for guiding experimentation, interpreting experiments, and increasing the efficiency of the experimental effort in obtaining the required understanding of proposed alloy system properties and performance. Modeling of microstructure evolution under irradiation has the potential to improve understanding of the response of various alloy systems to the higher temperature, higher dose conditions. Similarly, the performance of fuels in Generation IV reactors will depend on the synergistic interactions of complex phenomena that affect fuel lifetime and behavior. Understanding of these phenomena, and assessing the limits to be placed on fuel operation to ensure safety and reliability, will require the development of mechanistic models and their incorporation in fuel performance codes.

The development of models that can cut across the various scales, multi-scale modeling, is an important tool in promoting understanding of alloy behavior. Multi-scale modeling extends from electronic or atomistic scale models to molecular level models, to microstructure models to models for bulk mechanical behavior. Efforts to couple models across the various scales are being made in the reactor pressure vessel community and offer the promise of solving problems that are impossible to solve when confined to a single-length scale. Because the material challenges in the six Generation IV systems are largely the result of alterations in macroscopic properties (creep, fracture toughness, swelling) induced by atomistic changes (irradiation-induced point defects), alloy design in all concepts will benefit from modeling activities that link the length and property scales. Therefore, a single, or coordinated, Generation IV fuels and materials modeling effort is proposed.

3.10 Joining Techniques and Non-Destructive Evaluation

As the classes of materials proposed for Generation IV application are composed of a conglomerate of austenitic alloys, ferritic-martensitic alloys, ceramics, ODS materials and precipitation-hardenable Ni-base alloys, it shall be noted that there is a certain difficulty to join component parts during the fabrication of components. For some material classes, a suitable joining technique is not yet developed. Possibly, some of these materials, though of different compositions or form, will present similar challenges. It is quite possible that the application of effective techniques to one material or class of materials can be extended to other materials. Therefore, an R&D task to develop broadly applicable joining techniques for Generation IV application is proposed.

The possible application of materials, such as ODS alloys and ceramics, generates the requirement to establish suitable techniques for non-destructive examination because well-established methods that fulfill the requirements of nuclear application do not exist. In industrial practice, semi-finished parts and joints will need to be examined. Therefore, an R&D task to develop broadly applicable non-destructive examination techniques for Generation IV application is proposed.

3.11 Establishment of Codes and Standards

Because Generation IV concepts will require deployment of materials and components operating under unprecedented conditions, new codes and standards must be established to govern their use. Materials composition and property data that are collected during the development of Generation IV technologies must be obtained in accordance with quality assurance standards so that it can provide the necessary bases for codes and standards and for system licensing. Each participating country must provide experts to collectively guide the data collection and maintenance processes and to draft appropriate codes and standards.

4. FUELS AND MATERIALS CROSSCUTTING R&D RECOMMENDATIONS

The FMCG is providing some recommendations to guide the implementation of some of the R&D tasks proposed by the Technical Working Groups. In addition, the FMCG is proposing a specific set of R&D tasks for each of the identified opportunities described above. The recommendations and proposed R&D tasks are listed in this section. Although the detail supplied is modest, the descriptions are sufficient to define the objectives and nature of the activities and to provide a basis for estimating resources necessary for each task.

4.1 Structural Materials Properties and Behavior

4.1.1 Recommendation F&M: SMP&B

The inability of technologists to confidently propose materials for many of the Generation IV applications indicates that multiple materials, or classes of materials, will necessarily be considered. To reduce the scope of more expensive irradiation testing, pre-selection of candidate materials is recommended. The criteria for the pre-selection phase should be determined by a representative group of experts, and the pre-selection should result in a list of materials to be investigated using the first sets of irradiation properties tests in Tasks F&M: SMP&B 3 and 4 described below.

Testing of structural materials and fuels is likely to occur at several laboratory and test reactor locations throughout the U.S. and around the world. In order to ensure that data are readily accepted by companion laboratories and by the design community, and to avoid the unnecessary and costly duplication of experiments, a common set of testing and measurement technologies must be followed. These guidelines apply to testing of unirradiated material, testing of irradiated material in hot cells, and in-pile testing of fuels and materials in reactors. In many cases, standardized test methods have been developed by organizations such as the American Society for Testing and Materials. However, there are a number of testing procedures that will be critical to the assessment of properties for Generation IV application, for which no such standardization exists. For example, the method of measurement of crack growth rate in compact tension samples or the determination of cracking susceptibility using round bar samples in slow strain rate tests are without detailed, accepted procedures. In these cases, it is recommended that the Generation IV community will need to establish a set of common procedures and to consider round robin experiments to define the details of those procedures.

4.1.2 Tasks

Task F&M: SMP&B 1 Properties of Unirradiated Materials–*Estimated Cost*: $15M (over 8 years)

Objective: Determine mechanical and corrosion properties of alloys proposed for Generation IV reactor core components in the unirradiated condition.

Description: Conduct measurements of flow behavior (strength and ductility), toughness, fracture, fatigue, creep, creep-fatigue corrosion, and stress corrosion cracking on candidate alloys at the temperatures and stress levels anticipated for use in core components.

Duration: 2003–2011

Task F&M: SMP&B 2 Stability of Irradiated Microstructures—*Estimated Cost*: $50M (over 15 years)

Objective: Establish microstructural/microchemical stability of irradiated primary candidate alloys under consideration for Generation IV reactor applications.

Description: Conduct irradiations and measurements of the evolution of the dislocation microstructure, void and bubble microstructure, precipitate formation, dissolution and stability and radiation-induced segregation in candidate alloys at doses, dose rates, and temperatures expected for the Generation IV concept designs.

Duration: 2003–2018

Task F&M: SMP&B 3 Properties of Irradiated Materials—*Estimated Cost*: $15M (over 11 years)

Objective: Determine the mechanical and corrosion behavior of irradiated structural materials.

Description: Conduct mechanical testing and corrosion and stress corrosion cracking experiments on samples pre-irradiated in thermal neutron, fast neutron, or ion beam facilities capable of simulating the irradiation conditions. The test program would be conducted in a post-irradiation mode.

Objective: Determine fuel properties required for performance R&D.

Duration: 2005–2015

Task F&M: SMP&B 4 SCC Behavior—*Estimated Cost*: $15M (over 2 years)

Objective: Determine in situ mechanical behavior and corrosion/stress corrosion cracking (SCC) behavior of candidate materials for Generation IV structural materials.

Description: Conduct in-pile tests in reactor facilities to assess the mechanical (creep, fracture toughness, fatigue) and corrosion/SCC behavior of candidate alloys under irradiation in prototypical conditions.

Duration: 2010–2015

4.2 Fuel Materials Properties

Task F&M: FMP 1 Preparation for Fuel Properties Measurement—*Estimated Cost*: $5M (over 1 year)

Objective: Prepare for cost-effective measurement of fuel properties using combined resources of the Generation IV participants.

Description: Establish laboratories within the Generation IV participating countries for measuring key thermophysical properties and assessing phase equilibria for selected Generation IV fuel forms and compositions.

Duration: October 2002–September 2003

Task F&M: FMP 2 Selected Fuel Properties Measurement—*Estimated Cost*: $10M (over 2 years)

Objective: Determine fuel properties required for viability R&D.

Description: Measure thermal conductivity (or thermal diffusivity), enthalpy, and melting (or dissociation) temperature, all at selected temperatures, for selected Generation IV fuel forms with varying minor actinide additions.

Duration: October 2003–September 2005

Task F&M: FMP 3 Fuel Properties Studies—*Estimated Cost*: $15M (over 2 years)

Description: Measure thermal conductivity (or thermal diffusivity), enthalpy, and melting (or dissociation) temperature, as a function of temperature, and assess phase equilibria and diffusion behavior for relevant Generation IV fuel forms with a range of minor actinide additions.

Duration: October 2005–September 2007

4.3 Irradiation Testing

Task F&M:IT 1 Irradiation Test Capability—*Estimated Cost*: $10M (over 2 years)

Objective: Establish irradiation-testing capability necessary to support Generation IV concepts.

Description: Design and construct test vehicles for fast-spectrum testing of fuel and/or material samples in Joyo (Japan) and in a spectrum-enhanced position in the Advanced Test Reactor (U.S.). This capability would provide desired neutron spectrum and irradiation temperatures, but would not necessarily provide prototypic coolant flow and chemistry. Irradiation testing capability will also include ion irradiation facilities for rapid, low-cost screening evaluation of materials for core internal structures.

Duration: October 2003–September 2004

Task F&M:IT 2 Establish Irradiation Test Loops—Estimated Cost: $50M (over 4 years)

Objective: Establish capability for irradiation testing of fuels and materials under prototypic conditions, as necessary, to support Generation IV concepts.

Description: Design and construct test loops for use in thermal-spectrum test reactors to be determined in Joyo (Japan) and in a spectrum-enhanced position in the Advanced Test Reactor (U.S.). Test loops for each type of Generation IV coolant (supercritical water, helium, sodium, and Pb/Pb-Bi) will be designed to provide prototypic flow rates, channel temperatures, and coolant chemistry.

Duration: October 2003–September 2007

Task F&M:IT 3 Materials Sample Irradiation—Estimated Cost: $30M (over 4 years)

Objective: Provide materials samples with accumulated fast neutron exposure to support viability R&D investigations.

Description: Irradiate materials samples in a fast neutron test vehicle at a range of temperatures and to a range of exposures. Simulations using energetic light ions will also compliment the use of fast neutrons. These samples will be used for the investigations of microstructure and properties that are described in Section 4 above.

Duration: October 2002–September 2008

4.4 Reactor Fuel Transient Testing

Task F&M: TT Transient Test Capability—Estimated Cost: $40M (over 3 years)

Objective: Establish capability for transient testing of fuels under prototypic conditions.

Description: Design and construct instrumented test vehicles and test loops for use in transient test reactors such as TREAT (U.S.), the NSRR (Japan), or Cabri (France). Test loops for each type of Generation IV coolant (supercritical water, helium, sodium, and Pb/Pb-Bi) will be designed to provide desired flow rates, channel temperatures, and coolant chemistry to study fuel transient response and behavior under design-basis accident conditions.

Duration: October 2003–September 2006

4.5 Fuel Fabrication Technique Development

Task F&M: FD 1 Simplified Pelletizing Method—*Estimated Cost*: No incremental funds proposed

Objective: Develop the Simplified Pelletizing Method for application to all ceramic fuels under consideration for Generation IV concepts.

Description: The FMCG recommends that the Simplified Pelletizing Method be developed in accordance with the R&D tasks proposed by Technical Working Group 3. A broader range of pellet requirements should be considered, encompassing those for all relevant Generation IV concepts.

Task F&M: FD 2 Pyroprocess Materials Compatibility—Estimated Cost: No incremental funds proposed

Objective: Select materials that are suitable for use with molten alloys present in a pyrometallurgical recycle process.

Description: The FMCG recommends that R&D tasks identified by Technical Working Group 3 and the Fuel Cycle Crosscut Group be supported with an understanding that pyrometallurgical recycle may have application beyond the sodium-cooled reactor concept.

Task F&M: FD 3 Vi-pac Fabrication Parameters—Estimated Cost: $15M (over 4 years)

Objective: Develop parameters for vi-pac fabrication of ceramic fuels (oxide and nitride) under consideration for Generation IV.

Description: Determine the vi-pac fuel characteristics most likely to provide acceptable fuel performance for oxide and nitride vi-pac fuels of the compositions of interest for Generation IV systems.

Evaluate the necessary particle size distributions and vibrational compaction parameters necessary to produce the desired characteristics. Fabricate vi-pac fuel samples for irradiation testing.

Duration: October 2003–September 2007

4.6 Process Materials Selection and Development

No additional R&D tasks are recommended for this topic.

4.7 Materials Phenomena Modeling

Task F&M: MM 1 Radiation Effects Modeling—Estimated Cost: $10M (over 3 years)

Objective: Develop radiation effects models that will improve the understanding of irradiation performance of high-temperature materials to be investigated for Generation IV application.

Description: Multi-scale modeling techniques will be applied to radiation damage mechanisms that produce radiation effects of interest for Generation IV application. The effort will begin with an assessment of previous work that can applied to this task and will follow with development of techniques and approaches that can be broadly applied to different Generation IV materials applications. As the work progresses, further recommendations will be made as to whether a general modeling effort will meet the specific needs for each Generation IV concept or whether more specific development activities are necessary.

Duration: October 2004–September 2007

4.8 Joining Techniques and Non-Destructive Evaluation

Task F&M: J&ND 1 Joining Techniques—Estimated Cost: $15M (over 4 years)

Objective: Establish joining techniques that can be applied to Generation IV candidate materials.

Description: Adapt or develop, as necessary, joining techniques that allow fabrication of reactor components from materials with high melting temperatures, multi-phase alloys, or materials that form undesirable phases upon solidification during conventional welding processes. This task will begin with a review of industrial experience and will continue with technique-specific development work to be determined.

Duration: October 2006–September 2010

Task F&M: J&ND 2 NDE Techniques—Estimated Cost: $15M (over 10 years)

Objective: Establish non-destructive inspection techniques for Generation IV application.

Description: Challenges to the use of well-established non-destructive inspection techniques for Generation IV applications will be assessed and catalogued. Techniques that are currently available, but not necessarily used in nuclear applications, will be assessed for Generation IV application, and specific requirements will be identified. Adaptation or development of techniques for specific inspection needs will be completed.

Duration: October 2006–September 2016

4.9 Codes and Standards

Task F&M: C&S Establish Codes and Standards—Estimated Cost: $20M (over 22 years)

Objective: Ensure that standards and codes are established for deployment of materials in Generation IV systems.

Description: Each participating country will supply an expert to form a committee that will review the need for standards and codes to govern the deployment of materials in Generation IV systems. The committee will, over the course of Generation IV development, specify the criteria and characteristics that will be governed in the codes and standards, which will be used (as appropriate) in each participating country. The committee will also review the quality assurance methods being applied to collection and maintenance of fuels and materials R&D to ensure that subsequent data requirements for standards, codes, and licensing will be met.

Duration: October 2004–September 2025

5. FACILITY REQUIREMENT TO SUPPORT F&M R&D

A review of the R&D tasks proposed in the preceding section and in the R&D Scope Reports indicates that specialized R&D facilities will be necessary. In particular, fuels and materials R&D requires facilities (such as test reactors) capable of providing specific irradiation environments, shielded hot cells for examination and testing of irradiated fuels and materials, instruments for characterizing and testing fuels and materials, and test apparatuses for conducting specific tests of fuel rods or materials samples (e.g., transient heating furnaces located in shielded hot cells). Many of the nations that comprise the Generation IV International Forum and that are participating in the preparation of the Roadmap have such facilities that could be made available.

5.1 Irradiation Testing Facilities

A key facility requirement to support Generation IV fuels and materials R&D is associated with irradiation testing. Preparing a licensing application for a Generation IV system will require irradiation performance data and will be obtained by testing fuel designs in prototypical environments. Environmental conditions of interest include neutron spectrum, heat generation density, fuel and cladding temperature, coolant chemistry, and coolant flow rate. Irradiation temperatures and coolant conditions are typically obtained by experiment design, often requiring insertion of self-contained test loops into a test reactor. However, because the degree of radiation damage induced in materials under irradiation is due primarily to higher energy neutrons, the neutron spectrum is an important characteristic of an irradiation test and the spectrum of the test reactor typically cannot be altered significantly for an individual experiment. Therefore, selection of a test reactor includes consideration of its prevalent neutron spectrum—if the spectrum cannot be tailored as desired, then interpretation of the experimental results necessarily includes careful consideration of spectrum-dependent effects. In addition, the intention for several Generation IV systems to operate at relatively high temperature may challenge current test designs for test hardware, and obtaining desired temperatures will be more challenging if test temperatures significantly exceed those that are prevalent in the test reactor.

Thermal spectrum test reactors exist and appear to be available to support Generation IV irradiation testing requirements. However, there is a small number of fast-spectrum test reactors, and that number will be smaller still as the Generation IV program continues. The Joyo reactor in Japan and the BOR-60 reactor (or the planned BOR-60 replacement) in Russia are the only two fast-spectrum test reactors that will continue to operate beyond the next several years. The Phenix reactor in France is scheduled to be shut down after an additional 760 effective full-power days. The Fast Flux Test Facility in the U.S. is the facility best suited for the irradiation work described in the Generation IV R&D Scope Reports (in fact, it was designed specifically for irradiation testing and will accommodate test loops of different coolants). However, the Fast Flux Test Facility remains shutdown and is scheduled for irreversible shutdown activities later in this U.S. fiscal year.

Neutron irradiations are essential to evaluate and qualify materials for Generation IV systems. However, much can be gained in the understanding of neutron irradiation effects using ion-beam facilities. Currently, the DOE Office of Nuclear Energy, Science and Technology (DOE-NE) is sponsoring work using ion-beam facilities at both the University of Michigan and the Pacific Northwest National Laboratory. These facilities provide excellent capabilities for studying microstructural and microchemical changes during irradiation as well as corrosion and mechanical properties in many environments. The higher dose rates must be taken into account and the depth of penetration is typically not sufficient to assess bulk mechanical properties. Yet, charged particle irradiations can provide a low-cost method for conducting valuable radiation effects research in the absence of, or as a precursor to, verification experiments in reactors. While these facilities possess the capability to study radiation effects on modest-size programs, larger and more versatile facilities will be required for a major alloy design program such as that anticipated for Gen IV core internal materials.

5.2 Transient Testing Facilities

The types of transient tests conducted in the development of new fuel forms and in the safety assessment of reactor designs typically address overpower and undercooling conditions. Because these tests necessarily require conditions that are outside normal operating conditions for steady-state test reactors, specialized facilities are used to provide the desired test environment. Most transient test reactors are designed to provide overpower testing conditions, requiring the ability to safely simulate a nuclear reactivity insertion and associated power excursion for a test package, without jeopardizing the safety of the test reactor. Because such reactors are typically configured for testing of fuel materials in separate test loops that undergo temperature excursions, such reactors and loops are also employed to undercooling tests. Undercooling tests are different in that generated power in the fuel does not increase, but reduction in coolant flow rates induces higher temperatures within the test fuel.

Quite often, the conditions desired in an undercooling transient test can be simulated without nuclear heat generation. This is particularly true if the response, or the characteristic of interest to the test objective, is not dependent on temperature distribution through the fuel. For such cases, the transient test can be performed using fuel that is prototypic (e.g., irradiated with an as-irradiated microstructure and fission gas bubble morphology) and heated in a furnace located in a shielded hot cell.

5.3 Hot Cell Facilities and Special Experiment Apparatus

Two types of facilities are important to consider for material property determination, hot cell facilities equipped for materials property determination and specialized facilities that are located outside of hot cells, but which provide critical capability for material property determination:

1. **Hot cell facilities for testing of irradiated material**. This would include hot cells that are capable of testing irradiated materials at either the proper temperature range or in the proper environment or both. It is likely that the latter is rare and may ultimately limit the advancement of materials studies. For example, there are no facilities in the U.S. for testing SCC or mechanical testing of irradiated materials in Pb-Bi or supercritical water.

2. **Specialized facilities for testing of unirradiated material**. This would include facilities for simulating the proper environment for corrosion, creep, or stress corrosion cracking studies in Pb-Bi, Na, or supercritical water. While not in hot cells, these facilities are extensive, unique, and rare and may ultimately limit the advancement of materials studies for these new environments. For example, there are only three facilities (and two remain under construction) in the U.S. for testing SCC of alloys in supercritical water.

Appendix A

Higher Temperature Reactor Materials Workshop

Argonne National Laboratory, with facilities in the states of Illinois and Idaho, is owned by the United States Government and operated by The University of Chicago under the provisions of a contract with the Department of Energy.

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Higher Temperature Reactor Materials Workshop

Sponsored by the Department of Energy
Office of Nuclear Energy, Science, and Technology (NE)
and the Office of Basic Energy Sciences (BES)

March 18–21, 2002

La Jolla, CA

Todd Allen, Argonne National Laboratory-West
Steve Bruemmer, Pacific Northwest National Laboratory
John Elmer, Lawrence Livermore National Laboratory
Mike Kassner, Oregon State University
Arthur Motta, Pennsylvania State University
Robert Odette, University of California, Santa Barbara
Roger Stoller, Oak Ridge National Laboratory
Gary Was, University of Michigan
Wilhelm Wolfer, Lawrence Livermore National Laboratory
Steve Zinkle, Oak Ridge National Laboratory

June 2002

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Inspiration

"*We physicists can dream up and work out all the details of power reactors based on dozens of combinations of the essentials, but it is only a paper reactor until the metallurgist tells us whether it can be built and from what. Then only can one figure whether there is any hope that they can provide power."*

Dr. Norman Hilberry, former Director, Argonne National Laboratory

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FIGURES

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Appendix A

Higher Temperature Reactor Materials Workshop

SUMMARY

On March 18-21, 2002, the Department of Energy, Office of Nuclear Energy, Science, and Technology (NE) and the Office of Basic Energy Sciences (BES) sponsored a workshop to identify needs and opportunities for materials research aimed at performance improvements of structural materials in higher temperature reactors. The workshop focused discussion around the reactor concepts proposed as part of the Generation IV Nuclear Energy System Roadmap.

The goal of the Generation IV initiative is to make revolutionary improvements in nuclear energy system design in the areas of sustainability, economics, safety and reliability. The Generation IV Nuclear Energy Systems Roadmap working groups have identified operation at higher temperature as an important step in improving economic performance and providing a means for nuclear energy to support thermochemical production of hydrogen. However, the move to higher operating temperatures will require the development and qualification of advanced materials to perform in the more challenging environment. As part of the process of developing advanced materials for these reactor concepts, a fundamental understanding of materials behavior must be established and the data-base defining critical performance limitations of these materials under irradiation must be developed.

This workshop reviewed potential reactor designs and operating regimes, potential materials for application in high-temperature reactor environments, anticipated degradation mechanisms, and research necessary to understand and develop reactor materials capable of satisfactory performance while subject to irradiation damage at high temperature. The workshop brought together experts from the reactor materials and fundamental materials science communities to identify research and development needs and opportunities to provide optimum high temperature nuclear energy system structural materials.

Many materials R&D areas were identified to support development of Generation IV concepts. Based on the discussions at this workshop, the following areas appear to be the most critical for advancing Generation IV concepts.

- Research on advanced ferritic-martensitic and martensitic steels that allow for increased temperature of operation for liquid-metal and supercritical water concepts while improving toughness at lower temperatures. Issues include stability of oxides in ODS materials, basic microstructural and microchemical changes, and phase stability at high temperatures.

- Development and fundamental understanding of radiation performance of refractory alloys, ceramic composites, and coatings for high and very high temperature concepts.

- Research to improve radiation performance of austenitic stainless alloys including resistance to void swelling, embrittlement and stress corrosion cracking.

- Development of new high-temperature superalloys that are tailored for radiation environments (e.g., low nickel contents and controlled phase stability)

- Fundamental and applied understanding of the complexity of radiation damage in engineering alloys, including austenitic, ferritic, ferritic-martensitic, refractory metals, and ceramic materials.

- Fundamental and applied understanding of deformation and creep processes related to flow localization and grain boundaries.

- Fatigue in plants that are expected to load follow.

- Developing design data for nuclear graphite.

Although not specifically considered in this workshop, the attendees did note that significant corrosion related challenges in the areas of supercritical water, lead or lead-bismuth, and molten salts, as well as compatibility of fuels and cladding must be overcome before certain Generation IV concepts are viable.

Several fundamental science issues were identified:

- The co-evolution of all components of the microstructure, and their roles in the macroscopic response in terms of swelling, anisotropic growth, irradiation creep, and radiation-induced phase transformations should be studied within the of the science of complex systems.

- Displacement damage during irradiation creates a non-equilibrium, structure-chemistry evolution at the nanoscale and alters plasticity, corrosion and fracture processes. Fundamental understanding of these complex, interdependent, radiation-induced material changes is essential to underpin the development of Generation IV reactor systems.

- Key structural performance issues for most irradiated metallic alloys are time-independent embrittlement at low temperatures and time-dependent deformation and cracking at high temperatures. The evolution of non-equilibrium structures and chemistries promote a hardened matrix and lower grain boundary cohesive strengths thereby reducing the tensile stress required for cleavage or intergranular fracture. Advances in modeling and measuring the atomistics of fracture need to be combined with micromechanical models to better elucidate behavior in complex radiation-induced, multi-component nanostructures.

The workshop identified both physical research facilities and human resources as critical to supporting materials research for Generation IV concepts. Of specific note are the lack of a fast-spectrum irradiation facility and the expected near-term retirements of a significant number of experts without sufficient young scientists in training to replace the senior faculty, scientists, and engineers.

To control the workshop scope to a manageable level, the workshop did not address all of the issues that significantly weigh on the choice of materials for Generation IV systems. Specifically, the following items relative to structural materials were not addressed in detail:

- Chemical compatibility and corrosion issues

- Welding and joining

- Fuels, fuel-cladding compatibility, and fuel-recycle system compatibility

- Materials to minimize loss in recycle systems

- Material availability, cost, fabricability, joining technology

- Safety and waste disposal aspects (decay heat, etc.)

- Nuclear properties (neutron economy, solute burnup, etc.)

All of these issues play a critical role in determining if reactor concepts can be operated safely and reliably and at a reasonable cost. Because many of the proposed Generation IV concepts operate in unique coolants (e.g. lead, lead-bismuth, supercritical water, molten salt), research and development will be required to establish materials that can operate in these environments.

This workshop proved to be a useful initial discussion about the materials aspects of Generation IV nuclear energy systems. Because Generation IV reactor systems were not well enough defined at the time of the workshop to identify specific operating environment, the workshop took a rather broad view of materials issues. At the completion of the Generation IV Roadmap, the number of concepts being considered will be fewer than those considered at this workshop. Future workshops should aim to discuss a narrower issue in greater technical depth. Possible topics for future workshops include:

- Coolant specific corrosion and environmental cracking issues relative to specific Generation IV concepts (e.g., in lead-base, molten-salt, or supercritical water coolants)

- Materials to minimize process loss in fuel recycle systems

- Fuel development for specific Generation IV systems (e.g., nitride fuel development).

Compared to our knowledge of materials used in current light water reactors, the knowledge of in-reactor degradation of the materials being considered for Generation IV applications is significantly lacking. The demands of Generation IV are enormous, in terms of strength, toughness, resistance to corrosion, and dimensional stability, especially considering the synergistic operation of factors that stress the material and the associated failure mechanisms. If in the early 1960s reactor materials experts were asked to predict the problems that later occurred with UO_2 fuel, Zircaloy cladding, pressure vessel steels, stainless steels, and other nuclear energy system materials, they would have been unable to even conceive of the problems, let alone predict the outcome. This is especially true for complicated failure mechanisms such as irradiation-assisted stress corrosion cracking, which involve the synergistic interaction of various factors or the complex long term evolution of damage in pressure vessel steels. We are likely to discover similar failure mechanisms as we explore the extreme operation conditions of Generation IV reactors.

WORKSHOP

This report summarizes the discussions at the March 18–21 “Workshop on Higher Temperature Reactor Materials” sponsored by the Department of Energy Office of Nuclear Energy, Science, and Technology (NE) and the Office of Basic energy Sciences (BES) held in La Jolla, Ca. The report is based on the discussions of the technical experts who participated in the workshop, as summarized by the working group chairs. The workshop report is not based on an extensive search of the literature but rather on the expert opinion of the participants.

Participants

The following are the workshop participants.

Name	Organization
Allen, Todd	ANL
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Background

On March 18-21, 2002, the Department of Energy, Office of Nuclear Energy, Science, and Technology (NE) and the Office of Basic Energy Sciences (BES) sponsored a workshop to identify needs and opportunities for materials research aimed at performance improvements of structural materials in higher temperature reactors. The workshop focussed discussion around the reactor concepts proposed as part of the Generation IV Nuclear Energy System Roadmap.

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Generation IV Initiative

Beginning in 2000, the United States proposed to the Generation IV International Forum (GIF) that a technology roadmap be prepared to guide the Generation IV effort. The GIF is a group of ten countries that have joined together to advance concepts for a number of next-generation nuclear energy systems that can be licensed, constructed, and operated in a manner that will provide competitively priced and reliable energy products, while satisfactorily addressing nuclear safety, waste, proliferation and public perception concerns. The GIF agreed to support the preparation of a technology roadmap, and the roadmap became the focal point of their efforts.

As preparations for the technology roadmap began, it was necessary to establish goals for Generation IV nuclear energy systems. Eight goals for Generation IV are defined in three broad areas of economics, safety and reliability, and sustainability. Economics goals focus on creating a competitive life cycle and minimizing energy production costs and financial risk. Safety and reliability goals focus on safe and reliable operation, investment protection, and essentially eliminating the need for off-site emergency response. Sustainability goals focus on fuel utilization, waste management, and proliferation resistance.

The Generation IV initiative (http://gen-iv.ne.doe.gov/) has evaluated nuclear energy system concepts against the Generation IV Goals. The systems can be classified in four broad groups: water-cooled, gas-cooled, liquid metal-cooled, and non-classical systems. The concepts being evaluated at the time of this workshop are listed in Table 1. For those concepts that appear to have the best chance of meeting the Generation IV goals, detailed research and development (R&D) plans will be developed. The output of this workshop will support both concept selection and development of the R&D plans.

Table 1. Principal primary operating temperature ranges for Generation IV reactor concepts.

Low Temperature (<350°C)
• Integral primary system reactors • Simplified BWRs • Evolutionary pressure tube reactors • High-conversion LWRs
Intermediate Temperatures (~350–600°C)
• Supercritical LWRs-thermal and fast • Sodium-cooled LMRs • Lead/lead-bismuth cooled LMRs
Intermediate-to-High Temperatures (~600–900°C)
• Lead/lead-bismuth cooled LMRs • Molten salt fueled reactors • Prismatic gas-cooled reactors • Pebble bed gas-cooled reactors • Gas-cooled fast reactors
High Temperatures (>900°C)
• Very high temperature gas-cooled reactors • Molten salt cooled reactors • Gas fueled reactors

Design Needs-Mission Performance, Safety, Operability

Proper choices of cladding and structural materials are essential for the safe and reliable operation of any Generation IV system. The survivability of fuel cladding (prevention of cladding breach) must be ensured and predictable. Therefore, the wastage and strain of the cladding under all operating conditions must be understood. Cladding wastage can be caused by corrosion of the cladding by the coolant or chemical interaction between fuel or fission products. Cladding strain can be caused by fission gas or coolant pressurization, swelling of constrained components, or fuel cladding mechanical interaction. Structural materials must maintain adequate strength, toughness and ductility, must have corrosion rates that are acceptable, and must have adequate dimensional stability with regards to swelling and creep.

A wide range of structural materials are candidates for Generation IV applications, including austenitic stainless steels, Ni-and Fe-based super alloys, various grades of ferritic and ferritic-martensitic steels, oxide-dispersion-strengthened austenitic or ferritic steels, conventional high-temperature refractory alloys (based on V, Nb, Ta, Cr, Mo and W), and various composite materials (C/C, SiC/SiC, metal-matrix composites, etc.). Numerous factors must be considered in the selection of structural materials, including:

- Unirradiated mechanical and thermophysical properties
- Radiation effects (degradation of properties)

- Chemical compatibility and corrosion issues
- Material availability, cost, fabricability, joining technology
- Safety and waste disposal aspects (decay heat, etc.)
- Nuclear properties (neutron economy, burnup, etc.).

The workshop focused on assessing the first two criteria in this list, with particular emphasis on how these issues impact the allowable operating temperature and dose limits. Cursory information was also presented on the important area of chemical compatibility, but to maintain a manageable workshop scope, chemical compatibility was not a major focus of the workshop.

Because of the wide range of concepts being evaluated in the Generation IV Roadmap and the significant differences in the design maturity, the materials needs are not easily classified. To make recommendations to the Roadmap, the workshop participants divided the proposed concepts into four groups, based generally on the primary temperature range in which the concepts operate. These divisions are shown in Table 1.

Design Approach

Several factors define the allowable operating temperature window for structural and cladding alloys in a nuclear reactor. The lower operating temperature limit in all body-centered cubic (bcc) and many face-centered cubic (fcc) alloys is determined by radiation embrittlement (decrease in fracture toughness), which is generally most pronounced for irradiation temperatures below ~0.3 T_M where T_M is the melting temperature. Radiation-induced loss in toughness in bcc alloys at low temperatures (<0.3 T_M) is generally pronounced even for doses below 1 dpa. The loss of ductility in fcc materials is somewhat sensitive to damage rate. Some components that undergo a limited degree of radiation embrittlement may still be used in applications where the expected stress is small. The lower operating temperature limit for SiC/SiC composites will likely be determined by radiation-induced thermal conductivity degradation, which becomes more pronounced in ceramics with decreasing temperature. In addition, amorphization of SiC (which occurs at irradiation temperatures <150˚C) sets a firm lower temperature limit due to the 11% volume swelling.

The upper operating temperature limit of structural materials is determined by one of four factors, all of which become more pronounced with increasing exposure time: (1) thermal creep (grain boundary sliding or matrix diffusional creep), (2) high temperature He embrittlement of grain boundaries, (3) cavity swelling or anisotropic growth (particularly important for SiC and graphite, respectively), or (4) coolant compatibility/corrosion issues. In many cases, the upper temperature limit will be determined by coolant corrosion/ compatibility rather than by thermal creep or radiation effects. Based on existing irradiation data, void swelling is not anticipated to be a major concern in any of the bcc alloys up to damage levels in excess of 100 dpa, although further data are needed. Radiation-enhanced recrystallization (potentially important for stress-relieved Mo and W alloys) and radiation creep effects (due to a lack of data for the refractory alloys and SiC) need to be investigated.

An example of a stress-temperature design window is given in Figure 1 for Nb-1Zr[1]. The high-temperature limit is defined by thermal creep, the low-temperature limit by radiation embrittlement, and the upper stress limit by strength. For a specific application, coolant compatibility would also need to be factored into the analysis.

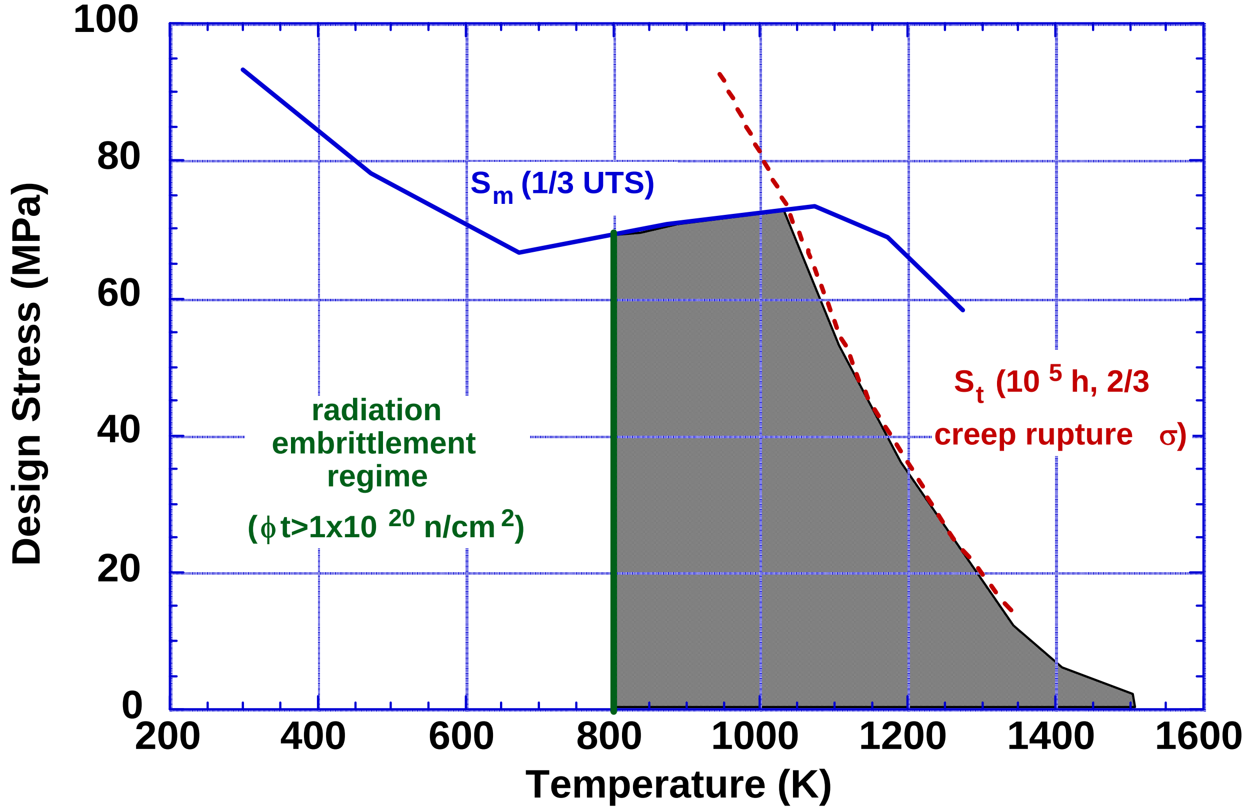
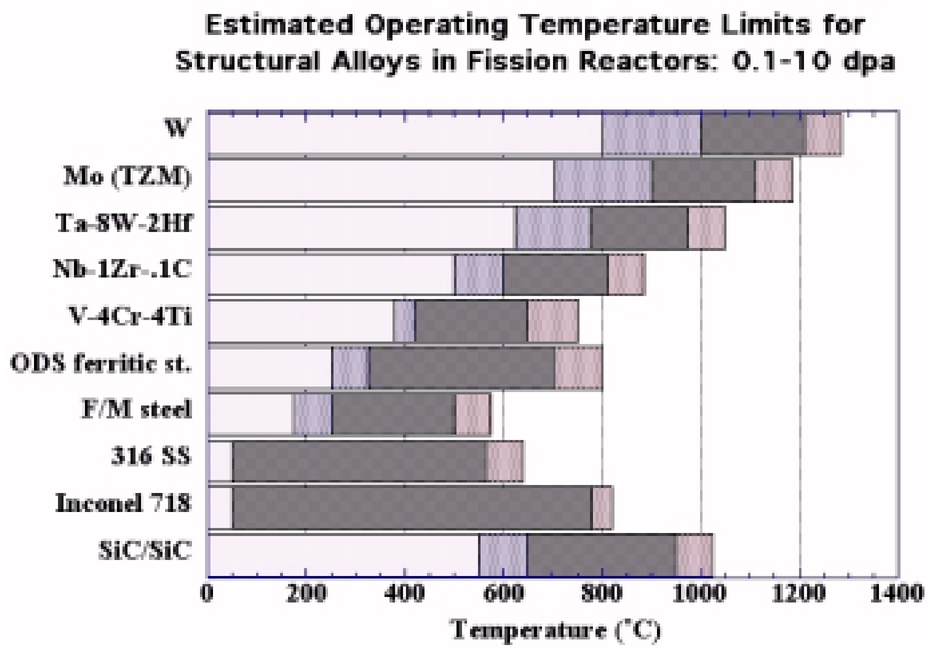


Figure 1. Stress-temperature design window for Nb-1Zr.[1]

Figure 2 summarizes the operating temperature windows (based on thermal creep and radiation damage considerations) for the several structural materials considered in the workshop[2,3]. Additional temperature restrictions associated with coolant compatibility need to be analyzed for specific reactor concepts. The specific values of the operating temperatures need to be combined with compatibility data for the candidate coolants to determine if the temperature window is reduced due to corrosion issues. As noted earlier, the minimum operating temperature for SiC/SiC is based on radiation-induced thermal conductivity degradation. The high temperature limit is based on thermal creep for all of the materials except SiC (void swelling was the limiting factor for SiC). A Stage II (steady-state) creep deformation limit of 1% in 1000 h (3×10^{-9} s^{-1} steady-state creep rate) for an applied stress of 150 MPa was used as an arbitrary metric for determining the upper temperature limit associated with thermal creep. Design-specific creep data would obviously be used to establish the temperature limits for longer times and lower stresses in several of the candidate materials.

Interaction between Radiation Damage and Microstructure

Radiation damage mechanisms occur in temperature ranges roughly defined by the homologous temperature, or fraction of the melting temperature. Figure 3 demonstrates the relationships. At low temperature, embrittlement due to radiation damage or due to the build-up of embrittling transmutation gases such as He and $^{3+}$H may cause a loss of toughness at low temperature. At intermediate temperatures, radiation creep and void swelling cause dimensional instabilities that must be understood for proper reactor operation. In addition, high-temperature helium embrittlement is likely unless the helium is properly managed. Reactor designs that increase operating temperatures above ~600°C will need to consider effectively strengthened alloys with higher melting temperature metals for structural



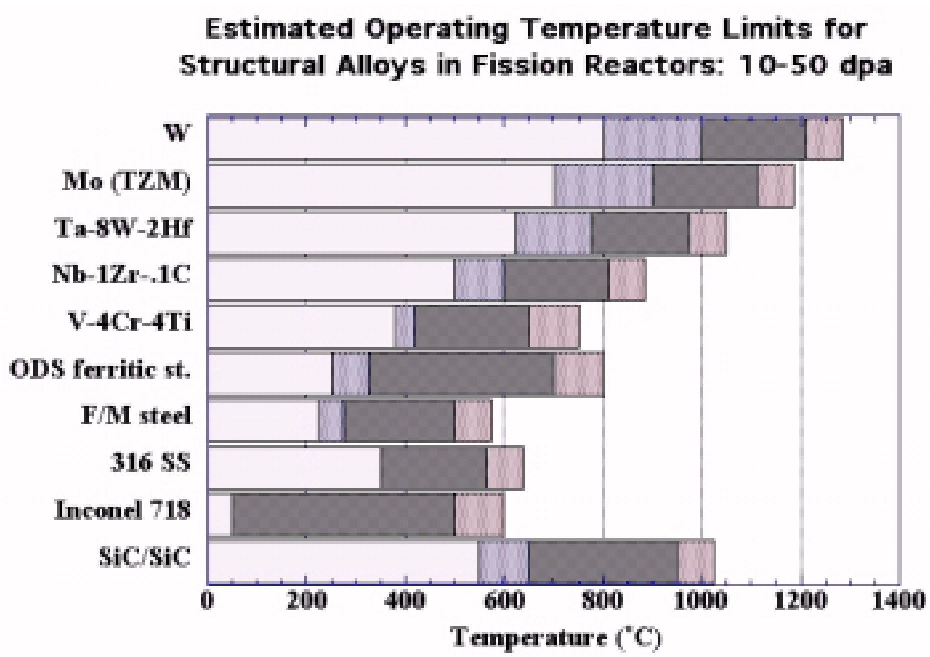


Figure 2. Operating temperature windows for various classes of reactor materials alloys. Top figure is for radiation doses up to 10 dpa. The second figure is for radiation doses up to 50 dpa. The upper and lower bands are temperature ranges where the materials performance may be adequate, but insufficient data currently exists to confirm performance.[1]

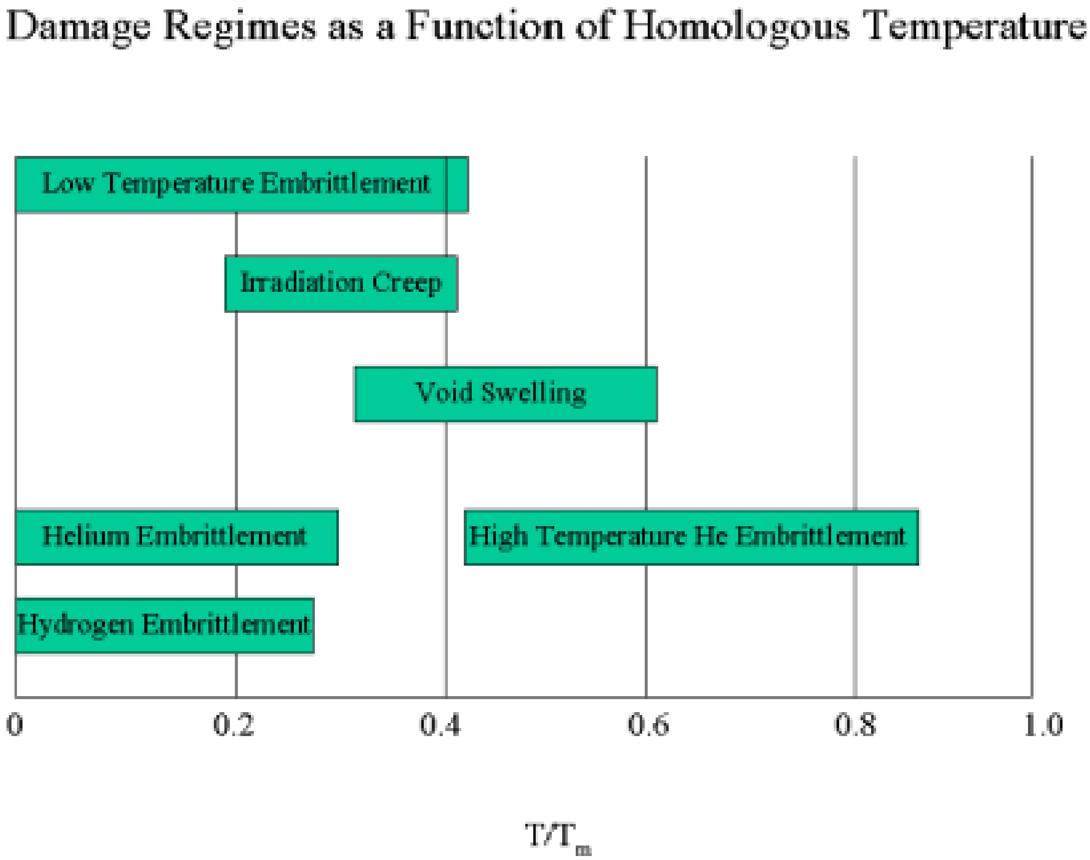


Figure 3. Temperature ranges over which radiation damage occurs.

components. Using these higher melting temperature alloys does not eliminate the possibility of similar radiation damage mechanisms as those found in construction materials (i.e., Fe- and Ni-base alloys) of current generation reactors. All of the higher temperature alloys need to be investigated to understand the effect of radiation damage and define performance limits in Generation IV reactor environments.

The changes in mechanical properties and dimensional stability in reactor components are caused by the development of microstructural features during irradiation. The complex relationships are illustrated in Figure 4. As an example, void swelling can cause unacceptable changes in dimension in reactor components. The density and size distribution of the voids is closely linked to the development of the dislocation loop structure as both voids and dislocations compete for point defects. Both void and dislocation development are affected by radiation-induced segregation and vice versa. The changes in composition in the area near the void and dislocations affect their growth and can induce precipitation. Similar complex relationships exist among all microstructural features and among microstructural features and bulk properties (e.g., strength, dimensional stability, ductility and toughness). In each of the following sections, the R&D challenges will be outlined for specific temperature regimes. Because of the strong relationship between bulk properties and microstructural features, each section will contain a description of the R&D challenges for both the bulk properties and the microstructure.

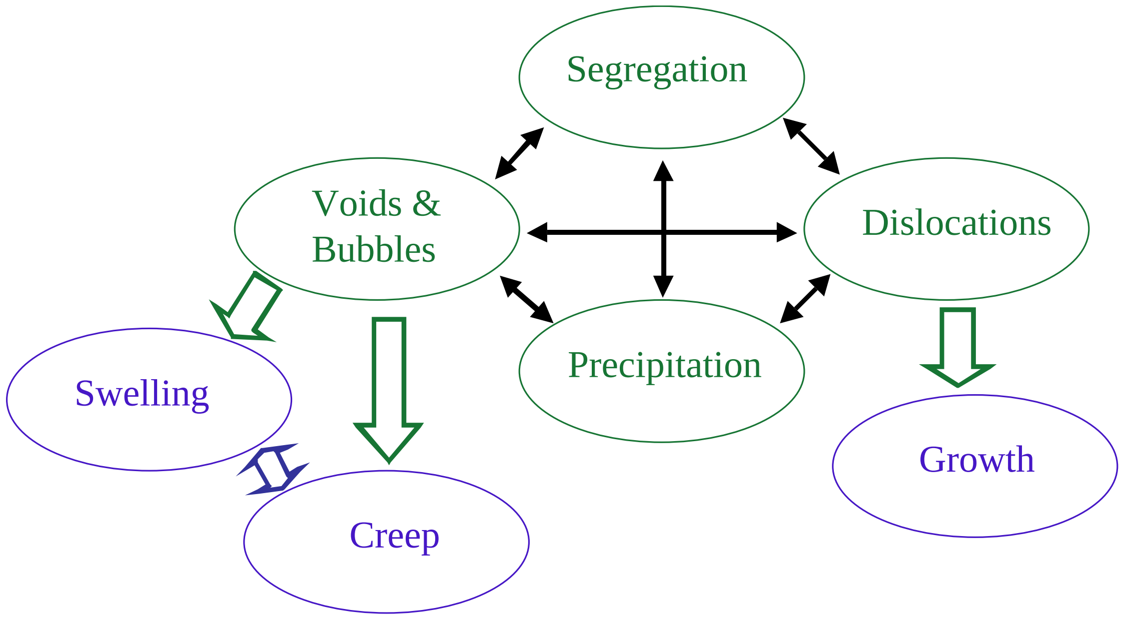


Figure 4. Relationship between microstructural features and dimensional stability.

Lower Temperature Reactor Issues (T< 350^{o}C)

This section discusses the materials R&D needs for components operating at temperatures less than 350^{o}C. As shown in Table 1, temperatures less than 350^{o}C are typically associated with water reactors (LWRs).

Materials issues for the low-temperature, advanced light-water-reactor (LWR) designs are far and away the best understood and manageable among the Generation IV concepts. Problems that are anticipated directly reflect economic and safety concerns being dealt with for existing Generation II plants (see Table 2).

High radiation levels in a reactor core produce changes in iron- and nickel-base austenitic stainless alloys leading to extensive hardening, a reduction in uniform ductility, and an increased susceptibility to intergranular stress corrosion cracking (SCC). This cracking process, called irradiation-assisted stress corrosion cracking (IASCC), is a serious concern for both boiling water reactors (BWRs) and pressurized water reactors (PWRs). Stainless steel components can become susceptible to cracking at doses less that 10% of the expected end-of-life dose and the likelihood of failure may increase with increasing service exposure. Susceptibility to IASCC is clearly linked to radiation-induced changes in the alloy microstructure and microchemistry, but fundamental understanding of controlling mechanisms has been elusive.

More extensive material changes can occur in components where gamma heating leads to temperatures greater than ~330^{o}C. Void swelling potentially becomes a life-limiting issue at these higher temperatures particularly for PWR designs where extremely high doses (>80 dpa) are reached. Swelling is not significant at temperatures below 300^{o}C due to the difficulty of nucleating and growing voids. Void nucleation and growth depend sensitively on the alloy composition, solute additions and He production rate. Radiation-induced precipitation can also occur at these higher temperatures in the matrix and at grain boundaries when the solubility limit for a particular solute is reached due to RIS. Matrix precipitation can further harden the alloy, while grain boundary phases can promote intergranular embrittlement in some cases.

Table 2. List of candidate materials and performance issues for low temperature (<~350°C) applications.

Structural Material	Performance Issues
Ferritic pressure vessel steels	Radiation embrittlement (toughness, DBTT*)*
Fe-base austenitic stainless steels	SCC, IASCC, high-dose embrittlement
Ni-base stainless alloys and superalloys	IGSCC, IG corrosion, weld metal SCC, IASCC
Zirconium alloys	Corrosion, hydriding
Ferritic/martensitic alloys e	Radiation embrittlement (toughness, DBTT), IGSCC, IASCC, hydrogen embrittlement

The microstructural development of austenitic iron- and nickel-base alloys under irradiation in the 270–350°C range is reasonably well known, but not always well understood. At temperatures below 300°C, the primary radiation-induced microstuctural components are small dislocation loops that promote hardening. The dislocation microstructure has been reasonably well characterized but poorly understood and not effectively modeled. Outstanding issues involve understanding the process of loop nucleation and the loop character. Radiation-induced segregation (RIS) occurs throughout the LWR temperature range and can cause significant composition variations at strong sinks such as grain boundaries. While segregation of the major alloying elements Fe, Cr, Ni is fairly well characterized and can be reasonably well modeled, the understanding of the behavior of minor elements that are believed to migrate as interstitials is poor and modeling capabilities are inadequate.

While improved mechanistic understanding and alloy development show potential for new austenitic alloys for advanced LWR applications, ferritic/martensitic alloys also have demonstrated better microstructure response while providing good strength with moderate corrosion resistance. These alloys are more resistant to swelling because of the long incubation period for void nucleation. Much of their recent development has been focused on improvements in fracture toughness after irradiation. However, there is very little data or understanding of other potentially important processes such as RIS, dislocation microstructure evolution or IASCC resistance under LWR conditions.

Significant levels of irradiation creep and associated stress relaxation can accumulate at LWR temperatures and can be accelerated by reactor transients in some cases. The current successful use of austenitic stainless steels suggests that they can be similarly applied in Generation IV reactors as long as the stress levels and duty cycles are comparable. Additional data is needed to determine the potential level of irradiation creep for materials other than austenitic stainless steels, and on transient effects in all materials. Current data indicates that the steady-state irradiation creep rate in ferritic-martensitic steels may be as little as 20% that of austenitic stainless steels. This needs to be confirmed, and transient creep rates need to be evaluated for the use of these steels as core structural materials.

Fatigue is not a significant issue in reactor internal components in current designs, although it must be considered when evaluating reactor piping. Potential sources of fatigue, such as flow-induced vibration are controlled or eliminated by design. Since LWRs in the U.S. are typically used to provide base-load power, major thermo-mechanical cycles are primarily associated with reactor startup and shutdown and the number of cycles is relatively small. If future plants are used for load-following, as is the case in some countries in Europe, the number of cycles may increase dramatically. In such a case, a more detailed design and material specific analysis for the impact of fatigue will be required. However, it is likely that existing fatigue design rules based on concepts such as traditional Wohler (S-N) curves or the more modern Coffin-Manson equation (an empirical curve of cyclic strain vs. lifetime) will be adequate for the application of most Generation IV reactor components that operate below 350°C.

As long as a component is expected to experience only a few hundred cycles before being taken out of service, the realm is that of low-cycle fatigue, and the lifetime can be predicted using the empirical Coffin-Manson equation. However, within the past few years, fatigue machines operating at 20kHz have been used to re-examine the concept of the endurance limit, and the findings indicate that there may be no endurance limit for failures in the very high cycle range. When new nuclear plants are designed, it will be important to incorporate this new knowledge for characterizing fatigue into the design and lifetime prediction process.

The effects of the reactor exposure environment and of radiation on fatigue in reactor internals warrant further research. Radiation-induced segregation could impact fatigue crack initiation and crack growth in a fashion similar to their impact on IASCC. The combined effects of RIS, radiation-induced hardening of the matrix, and the effects of water chemistry (e.g., due to radiolysis) could give rise to unexpected levels of fatigue damage accumulation.

Zirconium-based alloys are used for fuel cladding in all water-based designs and are well suited for use in this temperature range. They provide good corrosion resistance and adequate strength and are not prone to swelling. With proper alloy design, Zr alloys might be extended for use at slightly higher temperatures.

The main degradation mechanisms of Zr alloy cladding are corrosion and hydriding, but for the high temperature and burnup being considered, other mechanisms may become active. In particular, irradiation deformation mechanisms such as creep and growth may again become a consideration, as well as the onset of changes in rates of corrosion mechanisms (breakaway corrosion). The newer Zr alloys such as ZIRLO and M5, have much smaller rates of in-reactor degradation than Zircaloy-4 (meaning lower corrosion rates and lower hydrogen pickup and lower growth rates). At the current burnup limits (62 GWd/t fuel bundle average), these advanced alloys show corrosion rates (less than 20-30 micron oxides at the end of life) and irradiation growth rates that, extrapolated to high fluences would be quite tolerable. The uniform corrosion rates in BWR cladding are typically less than in PWR, but in BWRs accelerated localized corrosion (nodular corrosion or shadow corrosion) or hydriding problems are of more concern. Improved cladding (barrier cladding) and hydrogen water chemistry have helped address some of these problems

The very extensive experience base that currently exists, predicts good cladding behavior up to 10s of dpas and up to 5 years in-reactor under normal operation. The safety of fuel cladding at high burnup to accidents such as a reactivity insertion accident (RIA), a loss of coolant accident (LOCA) and an anticipated transient without scram (ATWS) has been recently evaluated and is now undergoing confirmatory research. Extrapolation to a factor of two is foreseeable within the current database. The effect of long-term (to 100 dpa) irradiation-induced microstructural changes on in-reactor degradation processes is not well understood. Processes such as precipitate dissolution, amorphization and formation of dislocation structures at high fluences might influence alloy behavior. For example, a transition in growth rates at intermediate fluences has been linked to the development of c-component dislocations, which have in turn been associated with stabilization of these loops by Fe released from precipitates that have undergone amorphization or dissolution.

The investigation of these processes that occur at high doses, coupled with high temperature corrosion testing could form the initial framework for research programs in high burnup, high temperature operation of Zr alloys.

The reactor pressure vessel remains perhaps the most important safety-related component in a nuclear power plant. Radiation-induced embrittlement of the ferritic steel is a critical concern particularly in the beltline region where long-term exposure to a moderate neutron flux leads to a significant reduction in fracture toughness. Considerable work has been performed establishing an empirical “master curve” approach to model the shift in the ductile-brittle transition temperature (DBTT) in ferritic pressure vessel steels. This research has helped quantify radiation-induced property changes and enable effective management of the degradation process. Fundamental links among alloy composition, radiation-induced microstructural evolution and embrittlement have improved alloy specifications and long-term properties. However, research is still needed to further reduce DBTT at higher doses, justify reduction in excess conservatism in reactor design and operation, and qualify remedial actions.

Corrosion and stress corrosion cracking are problems that have not been effectively solved for current LWR systems and remain an issue that must be recognized for Generation IV designs. These environmental degradation problems severely impact the economic operation of plants and, in a few cases, have created safety issues. The recent localized corrosion of the pressure vessel at the Davis Besse plant illustrates how potentially damaging these processes can be. Stress corrosion cracking continues to be a significant issue for nickel-base stainless alloys used for steam generator tubing, pressure vessel head penetrations and dissimilar metal weldments. The decades of LWR experience illustrates the need for underpinning science and the development of new alloys with improved corrosion and stress corrosion resistance in high-temperature water environments.

For low temperature reactors, the following issues are considered the most critical:

- Developing a mechanistic understanding of IASCC
- Determining the extent of void swelling in higher temperature components
- Understanding the complex, composition-dependent microstructural development that occurs in this temperature range
- Determining the feasibility of ferritic-martensitic steels for use in water reactor core internals
- The microstructural development and associated performance of zirconium alloys for use as fuel cladding to high burn-up
- An understanding of fatigue in plants that load-follow.

Improved radiation- and corrosion-resistant alloys are needed for the envisioned Generation IV water-cooled designs. Evolutionary modifications to current metallic alloys will likely be sufficient to achieve most performance goals. However, focused research and development activities are essential to establish basic understanding of degradation processes and build the necessary database to confirm alloy/component reliability in reactor service. The complex, interdependent evolution of radiation-microstructure and microchemistry must be better understood to produce alloys more resistant to degradation.

Intermediate Temperature Reactor Issues (350°C < T < 600°C)

This section discusses the materials R&D needs for components operating at temperatures between 350-600°C. As shown in Table 1, reactor concepts in this temperature range are supercritical water reactors and liquid metal reactors. Over this temperature range, candidate materials will require higher strength and greater resistance to diffusion-driven processes such as radiation-induced segregation and precipitation, void formation and growth, dislocation loop growth, creep, fatigue and high temperature corrosion and stress-induced corrosion cracking processes. Table 3 lists the prime candidate alloys and the associated performance issues addressed in this section of the report.

Table 3. List of candidate alloys and performance issues for intermediate temperature (~350–650°C) applications.

Structural Material	Performance Issues
Pressure vessel steels	Radiation embrittlement, (toughness, DBTT)
Fe-base austenitic stainless steels	Creep strength, swelling &embrittlement, corrosion, IASCC
Ni-base austenitic alloys and superalloys	He embrittlement, creep strength, swelling & embrittlement, corrosion, IGSCC, IASCC
Ferritic-Martensitic alloys	Radiation embrittlement (toughness, DBTT), corrosion, IASCC, hydrogen cracking, corrosion in led-based coolants and molten salts

The increase in reactor operating temperature and change in coolant environment alters material selection and many performance issues. Detrimental radiation-induced microstructural evolution (swelling and precipitation) and embrittlement severely limits the application of conventional austenitic stainless alloys. However, experimental swelling-resistant austenitic stainless steels have been produced for temperature up to ~600^{o}C during testing in advanced reactor programs.[4] Irradiation creep can be very severe in this temperature range for conventional austenitic stainless steels such as AISI-316 or AISI-304, and limits their use under high stresses to <600°C. Dispersed oxide precipitates or other second phase particles will be required for austenitic stainless alloys to maintain adequate strength at the upper end of the temperature range. The behavior of the precipitates under irradiation and their dose/temperature evolution is largely unknown, as is RIS, void swelling and dislocation microstructure interaction with the precipitates. The impact of an evolving precipitate structure on the dislocation microstructure, void nucleation and growth and RIS to interfaces are critical challenges. Very little data is available on these alloys and fabrication could well become a limiting factor.

At intermediate temperatures, helium diffusion and precipitation under irradiation will become more important than for present-day water reactor operating conditions. In this temperature range, the austenitic alloys pass through their peak in swelling, and at higher temperatures, void swelling will be minimal. The use of solute additions (e.g., Ti to stabilize carbon mobility and oversized substitutional solutes) may promote recombination and delay the onset of swelling so that doses of as much as 100 dpa may be reached before swelling becomes too large to accommodate by design. Helium generated from thermal neutron capture in Ni will migrate to grain boundaries and result in grain boundary bubble embrittlement. Here again, solute additions may be important in trapping He at vacancy-solute clusters to delay the aggregation of He into bubbles. RIS and irradiation-induced precipitation become increasingly important issues. RIS will peak in the 400–500°C range, while near the upper end of the range the high concentration of thermal vacancies will suppress RIS. The dislocation loop density will decrease sharply and the loop size will coarsen throughout the temperature range. At the upper end of the range, the microstructure should resemble an annealed condition with few loops and a low network density, with precipitation processes becoming increasingly important.

In this temperature regime, types and populations of all microstructure features change quickly with temperature. Little is understood about the complex interactions that could occur between microstructure features such as dislocations, voids/bubbles, RIS, precipitates when the characteristics of each is a very sensitive function of temperature. The interplay between these features and their relative sensitivities to temperature will be important to understand for austenitic Fe- and Ni-based alloy systems to be applied in this temperature regime. (see Figure 4)

The limitations with austenitic stainless alloys, especially void swelling, make the more radiation-tolerant ferritic-martensitic (F-M) steels attractive choices for high-dose core internal components.

Ferritic-martensitic alloys provide the potential to achieve doses above 200 dpa due to their inherent resistance to swelling. Primary concerns for these steels are the same as for pressure vessel steels, i.e., radiation or thermal aging effects on toughness and the DBTT. The creep rate tends to be significantly lower for ferritic and F-M steels than for austenitic stainless steels, but creep begins to become an issue at the top end of this temperature range (600°C). Recently, a class of ferritic and F-M steels have been developed in which a very fine (~1 to 4 nm diameter) oxide particles has been dispersed. The best of these oxide-dispersion strengthened (ODS) alloys have been shown to maintain this fine dispersion under thermal creep conditions of elevated temperature and stress. If this behavior is maintained under irradiation, the ODS steels may increase the upper temperature limit of the F-M steels by 100 to 200°C, and increase the operating stress limit in the 350 to 600°C temperature range. Limited irradiation data on a French ferritic ODS steel up to 600°C indicates that Chi phase formation can lead to crack nucleation at low plastic strains. This same alloy also showed evidence of oxide particle dissolution after irradiation to 80 dpa at ~500°C. There is virtually no experimental data on RIS in these systems.

Irradiation-assisted corrosion and IASCC are critical unknowns for the supercritical water reactor concept even though corrosion-resistant ferritic-martensitic alloys have been developed for fossil plants. Radiolysis effects may influence the base aggressiveness of the supercritical water environment with respect to corrosion and environment-induced cracking processes. In addition, radiation-induced material changes may alter properties and promote degradation in all of the possible alloy choices. The long-term corrosion and stress corrosion cracking behavior of ferritic-martensitic steels in lead-based coolants and molten salt is similarly, poorly known. These represent important areas of research requiring long-term tests on irradiated materials in supercritical water environments.

Both solid-solution and precipitate-strengthened nickel-base alloys have also been investigated in this temperature range. Solid solution alloys should have adequate strength and creep resistance up through the middle portion of the temperature range, and precipitation-hardened alloys are well suited to the upper end of the range. However, high He production and precipitation –induced grain boundary embrittlement will likely limit application in this temperature range to low (<20 dpa) dose applications. The high nickel content leads to the formation of high levels of helium from nuclear transmutation reactions initiated by thermal neutrons. Even relatively modest amounts of helium can significantly reduce ductility in these materials and may accelerate fatigue crack growth. Some nickel-based superalloys may be more resistant to embrittlement and creep deformation, but will likewise be limited by the formation of helium. Many of these alloys are also limited by softening and creep under irradiation above 500°C, and by the formation of brittle second phases. Nevertheless, precipitate stability and RIS in this system is not well understood, even in this low dose range.

Many of the Generation IV reactor designs produce less power than current LWRs and this lower power output may increase the likelihood that they will be used in a load-following mode. This would require more attention be given to fatigue and creep-fatigue interactions in this temperature regime. The required analysis methodology will depend on the cyclic loading frequency, absolute stress level, and temperature. The potential affect of RIS and exposure to the reactor coolant must also be considered. Thus, the analysis will be design and material specific. A reasonable database exists only for the austenitic stainless steels, and to a lesser extent, some of the advanced ferritic and F-M steels. Irradiation data is lacking on other potential alloy systems such as the ODS steels and high nickel alloys.

For intermediate temperature reactors, the following issues are considered the most critical:

- Research on advanced ferritic-martensitic and martensitic steels that allow for increased temperature of operation for liquid-metal and supercritical water concepts and improving toughness at lower temperatures. Issues include stability of oxides in ODS materials, basic microstructural and microchemical changes, and phase stability at high temperatures.

- Understanding the complex, composition-dependent microstructural development that occurs in this temperature range.

- Corrosion in supercritical water and lead-based coolants.

- Development of austenitic alloys resistant to swelling to very high dose.

High Temperature Reactor Issues (600°C < T < 900°C)

This section discusses the materials R&D needs for components operating at temperatures between 600-900°C. As shown in Table 1, gas reactors, molten-salt reactors, and lead-cooled metal reactors are expected to operate in this temperature range.

The ability to obtain higher thermal efficiency by increasing operating temperatures is limited by the range of possible structural materials. Table 4 summarizes candidate materials and some of the issues that may limit their application. Above 600ºC, the thermal mechanisms, rather than irradiation–induced mechanisms, dominate the behavior of most of the metallic alloy systems of interest. For example, the dominant creep mechanism becomes thermal, rather than irradiation creep. Conventional austenitic, ferritic, or ferritic-martensitic (F-M) steels cannot be used above 600°C with any significant level of applied stress. The best oxide-dispersion strengthened (ODS) F-M steels may have adequate creep strength for temperatures to ~800ºC, but, except for experimental heats, this has yet to be demonstrated. Moreover, there are essentially no data on the radiation stability of the oxide dispersion that is critical to the performance of these materials. The radiation- and aging-induced phase stability of these alloys will be critical to maintaining acceptable creep resistance, toughness, and a low DBTT. These ODS materials are promising but remain the subject of ongoing research. For example, a significant level of research is needed to understand the issues related to the oxide nanocluster formation and stability during the process of alloy fabrication by mechanical alloying and extrusion, and the mechanisms by which they influence high temperature plasticity.

The primary alternatives are materials such as precipitate-strengthened iron or nickel-base superalloys or refractory alloys of molybdenum, niobium, and tantalum. These are under consideration because of their high strength and good high-temperature creep resistance. Unfortunately, little is known about the microstructural development of either alloy class under irradiation at these temperatures. The degree of radiation-induced segregation (RIS) is not known but will likely be minimal due to the very high concentration of thermal vacancies that will tend to mitigate the effects of radiation-induced defect flow. The dislocation microstructure is likewise expected to be essentially free from irradiation-induced dislocation loops due to the instability of loops at such high temperatures. However, the effect of extremely high densities of small precipitates on both RIS and the dislocation microstructure in this temperature range is relatively unknown and little if any data exist. The stability of the precipitates themselves and their evolution with dose and temperature are unknown as well. Finally, the practical use of the refractory alloys is challenged by inherent toughness problems, limited understanding of radiation-induced material changes, a high sensitivity to oxygen-impurity pickup that can lead to further embrittlement, and cost.

While the nickel-base alloys may be used in components that are not exposed to neutrons, any in-core applications would have to be limited to very low doses due to transmutant helium production. RIS may further limit their nuclear applications by contributing to the formation of brittle phases with the potential for associated cracking. The upper temperature limit for out-of-core applications may be set by the thermal stability of the γ' phase that provides their creep strength. There is currently little creep or fatigue data in this temperature range for the refractory alloys. Substantial research is required before their

Table 4. List of candidate materials and performance issues for high temperature applications

Structural Material	Performance Issues
Iron and nickel-based superalloys	Creep behavior, Toughness, He Embrittlement
Ferritic-martensitic alloys	Creep behavior, Toughness, Radiation-induced embrittlement, Corrosion in lead-based coolants and molten salts, dispersion stability in ODS alloys
Refractory metal alloys	Creep behavior, Toughness, Radiation-induced embrittlement, Corrosion, Oxidation, Impurity pickup
Ceramic composites and coatings	Creep behavior, Radiation and environmental effects on interfaces, Toughness, Corrosion in lead-based coolants or molten salts
Graphite	Creep strength, Swelling, Toughness, Thermal conductivity

successful use can be anticipated. All issues related to microstructural stability and its impact on the accumulation of creep and fatigue damage need to be explored thermally as well as under irradiation, e.g., dislocation evolution, phase stability, and the possible impacts of enhanced diffusion- or radiation-induced segregation.

The importance of creep-fatigue interactions may increase at these high temperatures in components subjected to cyclic loads; little relevant data is available for any of the candidate materials. Creep rupture behavior of essentially all metals and alloys will be strongly influenced by the production of transmutant helium which will be mobile at high temperatures and is likely to accumulate in bubbles at grain boundaries. The level of concern is proportional to the helium production, which is highest in the nickel-base alloys. However, here again, there is not sufficient data to rank the candidate materials with respect to this issue.

Assuming that a reactor pressure vessel will be required for most Generation IV designs, the selection of an appropriate material for use in this high-temperature, thick-walled component will be difficult. Ferritic-martensitic steels may meet the strength requirements below about 700°C. However, they will be sensitive to radiation- and aging-induced degradation of fracture toughness. The higher operating temperature may limit the radiation-induced shift in the DBTT, but more experimental data and mechanical analysis is required before a pressure vessel material can be recommended in this temperature range. This analysis should include a design-specific review of anticipated accident conditions.

Compatibility between structural materials and coolants is a significant issue at these elevated temperatures. Corrosion and environmentally-induced cracking issues must be evaluated for the lead-cooled and molten-salt fueled reactor designs. Experience with Pb or Pb-Bi suggests that ferritic-martensitic steels may have adequate corrosion resistance depending on protective film formation, however more work is needed to establish this behavior. For the novel molten-salt fueled design, little is known concerning corrosion behavior in these aggressive fluoride salts. Although corrosion is not a primary issue for gas-cooled reactors, the impact of residual oxygen must be considered if oxygen-sensitive structural materials (such as the refractory alloys or SiC) are used.

At the upper end of this temperature range, ceramic composites may become a viable option for specific structural components. The only system that has been studied in some detail is SiC-SiC composites, for which a limited amount of radiation effects data is available because of the interest in this material by the fusion program. While the overall strength of the composite is related to the fiber strength, the strength and toughness under load depend on the integrity of the fiber-matrix interface. These interfaces can be degraded by radiation or environmental exposure, which leads to debonding. Fatigue data on the composites is preliminary but promising, particularly in comparison with monolithic ceramics. However, differential swelling between the fiber and matrix components can give rise to internal stresses as neutron exposure increases, leading to cracking at the fiber-matrix interface. For low oxygen partial pressures, crack growth is limited by either irradiation (T<1200°C) or thermal creep (T>1200°C) of the fibers. Significant levels of helium will be produced by nuclear transmutation in SiC, and this will exacerbate creep rupture behavior.

Graphite will be used as a neutron moderator and structural material in most thermal spectrum gas-cooled reactor designs. In spite of a lengthy history of research and use of graphite by the nuclear industry around the world, there is not now a qualified source for nuclear grade graphite in the U.S. Graphite is a composite material made from a coke filler, coal tar pitch binder and petroleum impregnating pitch. The material is formed, baked, impregnated and re-baked before being heated to temperatures in excess of 2500°C during the final phase of manufacture. Consequently, the graphite microstructure is complex, with both binder and filler derived graphite regions and porosity. Moreover, the method of forming (extrusion, molding or pressing) imparts structural anisotropy to the artifact because the filler coke and internal porosity are aligned during forming, i.e., the material develops texture.

The lack of design data for nuclear graphite is a critical issue. The graphite grades used in the past for reactors in Europe and the U.S.A. are no longer available. Moreover, these graphite grades cannot easily be recreated since the sources of filler coke (critical to graphite behavior) are no longer available. Consequently, new cokes and graphite grades will require qualification and an adequate design database must be developed. Developing and carrying out a program for qualification of nuclear graphite needs to be a near-term priority if gas-cooled reactor research is going to be seriously pursued. Other fundamental research needs for graphite include: characterizing primary damage in graphite, determining the fate of transmutant helium and its impact on microstructure evolution, and the effect of high doses of radiation on fracture toughness.

The dimensional changes occurring in the graphite during operation give rise to internal stresses. These stresses may be relieved by irradiation creep at temperatures far below those at which thermal creep is significant. Stress relaxation by irradiation creep has been studied and the mechanism is reasonably well understood. However, there is a paucity of relevant data for currently available graphite. Neutron irradiation damage causes a rapid initial increase in the strength and modulus of the graphite. However, structural changes that occur at higher damage doses eventually cause a reduction in strength and stiffness. Thermal properties such as thermal conductivity are also markedly affected by neutron irradiation. The displacement of carbon atoms from the graphite basal planes adds phonon scattering centers that in turn markedly reduce the phonon mean free path. Consequently, the thermal conductivity rapidly falls, by as much as an order of magnitude, at relatively small damage doses. Moreover, the effect is temperature dependent because at higher irradiation temperatures the displaced carbon atoms are more mobile and can recombine with lattice vacancies. Thus, the extent of thermal conductivity degradation is more marked at lower irradiation temperatures.

For high temperature reactors, the following issues are considered the most critical:

- Research on advanced ferritic-martensitic and martensitic steels that allow for increased temperature of operation for liquid-metal and supercritical water concepts and improving toughness at lower temperatures. Issues include stability of oxides in ODS materials, basic microstructural and microchemical changes, and phase stability at high temperatures.

- Understanding the complex, composition dependent microstructural development that occurs in this temperature range.

- Development and fundamental understanding of radiation performance of refractory alloys, ceramic composites, and coatings for high temperature concepts.

- Qualification of an appropriate material for use as a high-temperature, thick-walled pressure vessel.

- Developing design data for nuclear graphite.

Very High Temperature Reactor Issues (900°C < T)

This section discusses the materials R&D needs for components operating at temperatures greater than 900°C. As shown in Table 1, temperatures greater than 900°C are associated with very high temperature gas reactors, molten salt reactors, and vapor core reactors.

The phrase "here there be dragons" was written on maps to warn ancient mariners against venturing beyond the edge of the known world. The very high temperature operating environments proposed for some Generation IV reactor designs might be similarly demarcated. Most conventional metallic alloys, those for which a reasonable experimental database and broad operation experience exists, cannot be used in this temperature range where thermal creep and creep rupture are the limiting phenomena. All of the issues discussed above in the section on temperatures between 600 and 900ºC apply here also, with the emphasis proportionally increased. Possible alternate materials may have acceptable high-temperature performance but there are insufficient data to design complex engineering structures, particularly those that will be exposed to neutron radiation.

Of the potential metal-based systems, only tungsten- and molybdenum-based systems are believed to have the potential to operate in this temperature range. Little or no data exist on the development of microstructure under irradiation for either of these systems. There is virtually nothing known about void formation, swelling, RIS, dislocation microstructure or precipitate formation and stability under irradiation at temperatures above 900°C. Transmutation could become a more significant problem in some systems such as tungsten, where transmutation leads to the formation of rhenium that precipitates as a metallic phase. The extreme temperatures also present a problem in conducting controlled experiments in existing irradiation facilities.

Molybdenum-base and tungsten-base alloys may have sufficient creep strength at temperatures up to 1100 and 1200^{o}C, respectively. However, as noted in the previous section, these alloys tend to be inherently brittle and further degraded by irradiation exposure. Their sensitivity to further embrittlement is increased by exposure to oxygen. A similar problem has also been discovered for high-temperature intermetallics and monolithic ceramics. Experience from the aerospace industry indicates that the use of ceramic coatings may limit corrosion and impurity pickup, but there is no information on the stability of these coatings in an irradiation environment.

This leaves ceramic composites as possibly the best option for these very high-temperature applications. SiC-SiC would be a first choice for temperatures up to ~1000^{o}C since it is the only composite system for which even a limited amount of irradiation effects data are available. Initial results suggest that ceramic fibers with better high-temperature strength will be required to achieve higher operating temperatures. Concerns about the effect of helium produced by nuclear transmutation in SiC also increase as the temperature increases. If reactor designs with component temperatures in this range are to be seriously pursued, an effort must be made to identify and investigate a range of possible candidate ceramics and ceramic composites.

For high temperature reactors, the following issues are considered the most critical:

- Understanding the complex, composition-dependent microstructural development that occurs in this temperature range.

- Development and fundamental understanding of radiation performance of refractory alloys, ceramic composites, and coatings for high temperature concepts.

Welding and Joining

The workshop did not dedicate significant time to the discussion of welding and joining, but this topic is expected to be important for both unirradiated and irradiated materials used in Generation IV systems. As an example, one class of materials being discussed to allow increased reactor operating temperatures is oxide-dispersion strengthened alloys. These materials contain large amounts of oxide particles distributed on a very fine scale, and will present a major challenge for welding. Conventional arc, laser or electron beam welding techniques would require filler metals to join ODS alloys to prevent unwanted oxide/heat-source interactions that would occur during autogeneous welding of ODS alloys. Existing filler metals may not provide the exceptional high temperature properties of these ODS materials. Research to develop high-temperature ODS filler metals, or to investigate solid state joining methods such as friction stir welding or explosive cladding where the problems associated with melting ODS alloys using conventional welding techniques can be avoided would be useful. For other new materials developed under Generation IV, each is expected to have its own unique set of issues related to welding and joining.

Basic Science Issues

The intent of the Workshop was to identify needs and opportunities in materials research aimed at performance improvements of structural materials in high temperature reactors. The needs and opportunities identified were to span from fundamental mechanistic understanding of materials performance to application in nuclear energy systems. The recommendations on the most important fundamental science are described in this section.

Microstructural Development

The International Conference held at Albany in June of 1971 ushered in a new era of fundamental research in radiation effects in metals and alloys. It was motivated and guided by the surprising discovery of void swelling, a phenomenon that was unexpected at the high irradiation temperatures in nuclear power reactors. Since that time, an enormous amount of excellent research has been conducted on the microscopic nature of radiation effects at elevated temperature, at which the most basic defects, vacancies and interstitials, are mobile. Although most of the defects produced in the collision cascades annihilate and restore the material to its equilibrium state, it was realized that a subtle bias exists in this complex system driven continuously from its equilibrium state which is the ultimate cause of void swelling, irradiation creep, phase stability, and the change in mechanical properties. In addition, radiation-induced segregation of impurity and alloying elements to grain boundaries, dislocations, and voids can occur, and this in turn can trigger dissolution as well as formation of new phases and precipitates. Each of these radiation effects has become a topic of in-depth research and scientific inquiry, and each displays a richness of diverse behavior, depending on the material, the irradiation temperature, the dose, and even the dose rate.

Yet in spite of this bewildering complexity, there have also emerged similarities and universal features. For instance, all metals and alloys studied so far have exhibited void swelling when subjected to irradiation in the temperature range between about 0.3 to 0.65 of their respective melting points. Void swelling is preceded by an incubation period which can last from only 0.1 dpa to 100 dpa, followed by a steady-state swelling stage which may or may not merge into a saturation stage. In parallel with the incubation, dislocation loops form and grow, eventually merging with the network dislocations. The latter continues to evolve until a saturation density is reached which, at least in austenitic steels, has been found to be independent of the initial dislocation density.

Another phenomenon, namely irradiation-enhanced creep appears to be a universal radiation effect at temperatures below 0.65 of the melting point. In fact, it probably occurs at all temperature, but it becomes masked by thermal creep at higher temperatures.

Irradiation creep commences long before the onset of void swelling, yet it becomes closely coupled to void swelling. There are indications from breeder reactor experiments that irradiation creep may disappear when void swelling reaches its steady-state rate. At that point, the radiation-induced expansion rate even under tensile loads is limited by the expansion rate of un-stressed material.

The universality of the total rate of expansion is again a surprising discovery that was not supposed to occur. It cannot be understood from present theoretical models of void swelling and irradiation creep.

It appears that after 30 years of much detailed and in-depth research, <u>the sum of all radiation effects does not add up entirely to the full behavior of a material exposed to continuous irradiation at elevated temperatures</u>. This, as we now know, is the hallmark of the dynamic and kinetic response of a complex system when driven far from thermodynamic equilibrium. The detailed kinetic processes are both deterministic and stochastic, they are coupled, and may evolve into new kinetic processes as the microstucture of the system evolves.

The crucial components or elements of the microstructure that evolve with irradiation are voids and bubbles, dislocation loops and stacking fault tetrahedra, and network dislocations. In addition, in many types of steels, carbides and other precipitates may form during the incubation period for void nucleation and influence the evolution of the other three microstructural elements. Finally, the generation of nuclear reaction products introduces potent elements for assisting void nucleation and growth. In particular, the diffusion and segregation of helium and hydrogen to vacancy clusters and voids is a major ingredient in modeling void swelling.

The major scientific challenge in the field of radiation effects in solids is the co-evolution of all these components of the microstructure, and their roles in the macroscopic response in terms of swelling, anisotropic growth, irradiation creep, and radiation-induced phase transformations. The framework for this new era of radiation effects research should be the science of complex systems. While this emerging field is still in its definition phase, the new topic proposed for radiation effects in solids is well defined. It may in fact serve as a prime example or model of a complex system evolving in response to the continuous flow of energy which maintains the system in various states of non-equilibrium.

The fundamental basis for developing radiation-resistant materials for high-temperature reactor concepts must be established. For example in strengthened metallic alloys, critical understanding of the radiation stability of all phases is essential, taking into account the complex interactions taking place in the evolving system on the nanoscale. This requires mechanistic modeling of radiation damage processes in a wide range of metallic, intermetallic and ceramic phases plus the ability to model collective damage evolution in an integrated system. While a strong underpinning capability exists for many simple metals, the ability to assess behavior in complex alloys or in non-metallic systems is very limited.

Deformation and Embrittlement

Displacement damage during irradiation creates a non-equilibrium, structure-chemistry evolution at the nanoscale and alters plasticity, corrosion-oxidation and fracture processes. Fundamental understanding of these complex, interdependent, radiation-induced material changes is essential to underpin the development of Generation IV reactor systems. Remarkable advances over the last 20 years in measurement and modeling tools (driven in most cases by the Office of Basic Energy Sciences) enables a unique opportunity to establish the critical foundation for radiation-resistant materials development. Atomistic response defining structural behavior of irradiated materials can be interrogated for the first time.

Deformation and embrittlement issues at various temperature regimes critical to Generation IV reactor concepts were evaluated. In each temperature range, options for core structural materials were identified along with important performance limitations, materials development needs and core underpinning science issues.

Radiation-induced evolution of non-equilibrium structures and chemistries alter the basic deformation processes within the matrix and grain boundaries. Complex interactions among defects (clusters, loops, bubbles and voids), including elemental segregation, precipitation and dynamic dislocation processes are poorly understood. Key research has been initiated in isolated simple systems, but must be expanded to address fundamental issues controlling defect-defect interactions, flow localization through segregated defect nanostructures and interfacial deformation (dislocation emission, transmission and sliding).

The key structural performance issues for most irradiated metallic alloys are time-independent embrittlement at low temperatures and time-dependent cracking at high temperatures. The evolution of non-equilibrium structures and chemistries promote a hardened matrix and lower grain boundary cohesive strengths thereby reducing the tensile stress required for cleavage or intergranular fracture. At high temperatures, the radiation-induced changes in the matrix and particularly at grain boundaries can promote creep embrittlement. Irradiation provides a challenge and an opportunity to explore the basic processes controlling transgranular and intergranular processes. The competition between matrix and grain boundary processes is an integral part of deformation and fracture in both irradiated and unirradiated materials. Advances in modeling and measuring the atomistics of fracture need to be combined with micromechanical models to better elucidate behavior in complex radiation-induced, multi-component nanostructures.

Although corrosion issues were not directly assessed within the Workshop, environment-assisted cracking was identified as a primary materials issue for most reactor concepts. Underpinning research focused on the unique radiation-environment interactions that promote irradiation-assisted stress corrosion cracking and dynamic embrittlement is needed. Fundamental unknowns were recognized for radiation-enhanced corrosion and cracking in supercritical water and lead-based coolants.

High Temperature, Time Dependent Deformation and Fatigue

Investigation of non-equilibrium thermodynamics in driven systems and on the nanoscale is needed. Understanding this behavior has direct implications to the performance of several materials that may be used in Generation-IV reactors.

The presence of persistent point defect supersaturations in materials undergoing displacive irradiation give rise to unique phenomena, such as radiation-induced solute segregation and solute clustering. This environment provides a unique opportunity to study material behavior far from equilibrium.

The formation and evolution of nanoscale solute-oxide clusters in metals and alloys during mechanical alloying and subsequent thermo-mechanical treatment. These clusters appear to demonstrate non-equilibrium thermodynamic behavior, e.g. in their composition and diffuse interfaces.

Mechanisms influencing dislocation motion and plasticity in materials containing a finely-dispersed nanoscale second phase are not well understood. Cottrell-like atmospheres consisting of the same solutes that are contained in the nanophases have been observed. The relative importance of these atmospheres and the nanophases in strengthening these materials is not known.

Time-dependent, high-temperature plasticity associated with crack growth when both thermal creep and fatigue mechanisms are operative is not well understood. The effect of creep-fatigue interaction on the local microstructure in front of the crack tip, and the feedback due to microstructural changes may influence subsequent crack growth.

Cross Cutting R&D

Many materials R&D areas were identified to support development of Generation IV concepts. Table 5 lists the different concepts proposed for Generation IV and the expected degradation issues. In any situation with limited budgets, choices must be made regarding the research that is most effective at advancing the concepts. This research could be crosscutting and therefore applicable to many concepts or critical to the advancement of a specific concept. Based on the discussions at this workshop, the following areas appear to be the most critical for advancing Generation IV concepts.

- Research on advanced ferritic-martensitic and martensitic steels that allow for increased temperature of operation for liquid-metal and supercritical water concepts and improving toughness at lower temperatures. Issues include stability of oxides in ODS materials, basic microstructural and microchemical changes, and phase stability at high temperatures.

- Development and fundamental understanding of radiation performance of refractory alloys, ceramic composites, and coatings for high and very high temperature concepts.

- Research to improve radiation performance of austenitic stainless alloys including resistance to void swelling, embrittlement and stress corrosion cracking.

- Development of new high-temperature superalloys that are tailored for radiation environments (e.g., low nickel contents and controlled phase stability)

- Fundamental and applied understanding of the complexity of radiation damage in engineering alloys, including austenitic, ferritic, ferritic-martensitic, refractory, and ceramic materials.

- Fundamental and applied understanding of deformation and creep processes related to flow localization and grain boundaries.

- Fatigue in plants that are expected to load follow

- Developing design data for nuclear graphite

Although corrosion was not a specific consideration of this workshop, the attendees do noted that significant corrosion related challenges must be overcome before certain Generation IV concepts are viable.

- Corrosion in supercritical water

- Corrosion in lead or lead-bismuth

Table 5. Principal primary loop structural materials issues for Generations IV Reactor Concepts.

Concept Group	Coolant Temps (°C)	Principal Structural Materials Identified Other Structural Materials Probable	Principal Materials Issues Identified Other Key Issues?
Low Temperature (<350°C, Evolutionary LWR's + Some Unknowns)			
Simplified BWRs	215-300	Zr Alloy Cladding Ni-Base Alloy Steam Generator Tubes Ferritic Steel Vessel Austenitic Stainless Steel Core Internals	Time-dependent behavior at high burnup Zircaloy corrosion/hydriding at high burnup Fuel-side corrosion by (U,Th)O2 fuels IASCC, HIC, Prim/Sec. IGSCC, Vessel DBTT
Evolutionary Pressure Tube Reactors	310–330	Zr and Zr-Nb Alloy Cladding/Pressure Tube Ferritic Steel Vessel Stainless Steel (Unspecified) Pressure Tube	Fuel-side corrosion by (U,Th)O2 fuels Zircaloy corrosion/hydriding Internals Swelling, IASCC, Primary/Sec. IGSCC
Integral Primary System LWRs	270–330	Zr Alloy Cladding Ni-Base Alloy SG Tubes Ti-Base Alloy SG Tubes Ferritic Steel Vessel Austenitic Stainless Steel Core Internals	Time-dependent behavior at high burnup Zircaloy corrosion/hydriding at high burnup Fuel-side corrosion by burnable poisons Corrosion/erosion of Ti-alloy SG tubes Corrosion of in-vessel components Swelling, IASCC, IGSCC, Vessel DBTT
Pebble-Fueled PWR	300–350	UO2 + Zr Alloy Clad Fuel Structural Alloys (Unspecified) Austenitic SS Core Internals + Ferritic Steel PPV	Fabrication of Zr coatings on UO2 spheres Erosion/fretting of Zr coatings, corrosion/hydriding Internals Swelling, IASCC, Primary/Sec. IGSCC
Pebble-Fueled BWR	285–370	Layered Particulate Fuel Structural Alloys (Unspecified) Ferritic Steel Vessel Austenitic Stainless Steel Core Internals	Wear of components by SiC, Erosion/corrosion of SiC Diffusion of fission products through SiC Stability of fuel layers at high burnup Swelling, IASCC, IGSCC
High-Conversion LWRs	Unspecified (<350 ?)	Zircaloy Cladding Structural Alloys (Unspecified) Austenitic SS Internals, Ferritic Steel RPV	Irradiation behavior of Zr in hardened spectrum Fuel-side corrosion by (U,Th)O2 fuels
Passive Pressure Tube Reactor	Unspecified (<350 ?)	TRISO Fuel SiC Coatings on Moderator/Fuel Matrix Graphite Moderator/Fuel Matrix Zr Alloy Pressure Tube Ferritic Steel Vessel	Fabrication/inspection of TRISO fuel Diffusion of fission products through SiC Fabrication of structural SiC composites Time-dependent behavior of SiC coatings/composites Corrosion of SiC or SiC/SiC cladding in water

Table 5. (continued).

Concept Group	Coolant Temps (°C)	Principal Structural Materials Identified *Other Structural Materials Probable*	Principal Materials Issues Identified *Other Key Issues?*
Intermediate Temperatures (~350–600°C, Evolutionary from LWR/Fossil, FBR + Some Unknowns)			
Supercritical Thermal LWRs	375–500	Ni-Base Alloy Cladding *Ferritic RPV* *Ferritic SS Core Internals ?* *Different Cladding*	Swelling, He, Creep of Ni-base alloy cladding *Swelling & Embrittlement of core internals* *Corrosion/SCC/IASCC in supercritical water*
Pebble-Fueled Supercritical LWR	280–540	TRISO Fuel Structural Alloys (Unspecified) *Ferritic RPV* *Ferritic SS Core Internals ?*	Corrosion of SiC in boiling water Diffusion of fission products through SiC Erosion of SiC, wear of other alloys by SiC *Corrosion/SCC/IASCC in supercritical water*
Supercritical Thermal HWRs	350–625	Zr Alloy Cladding Ni-Base Alloy Cladding for High Temperature *Ferritic SS Core Internals ?*	Swelling, He, Creep of Ni-base alloy cladding *Swelling & Embrittlement of core internals* *Corrosion/SCC/IASCC in supercritical water*
Supercritical Fast LWRs	Unspecified	$ZrH_{1.7}$ Moderator (is it really fast?) Stainless Steel (Unspecified) Structures *Ferritic RPV* *Ferritic SS Core Internals ?*	Thermal & dimensional stability of $ZrH_{1.7}$ Barrier coatings to prevent H loss *Swelling, Creep & Embrittlement of core internals* *Corrosion/SCC/IASCC in supercritical water*
Sodium-Cooled LMRs	400–550	Many steels identified for components Ferritic High Cr and Ferritic/Martensitic Steels Austenitic Stainless Steels ODS Steels	*Swelling, Creep & Embrittlement of core internals* *Thermal or Radiation-Induced Toughness/DBTT* *Corrosion/cracking at higher temperatures*
Intermediate-to-High Temperatures (~600–900°C, Evolutionary + Many Critical Unknowns)			
Lead/Lead-Bismuth Cooled LMRs	400–700	Austenitic Steel High Cr Ferritic Steels Ferritic/Martensitic Steels ODS Steels Advanced High Temp Alloys	Creep, Swelling & Embrittlement of core internals Thermal or Radiation-Induced Toughness/DBTT Corrosion & environmental cracking, IASCC
Molten Salt Fueled Reactors	400–700	Graphite Structure Ni-Base Alloy & Ni-Mo Structures Advanced High Temp Alloys	Corrosion, environmental crackings in fluoride salts Swelling, Creep & He Embrittlement of Ni alloys Thermal or Radiation-Induced Toughness/DBTT

Table 5. (continued).

Concept Group	Coolant Temps (°C)	Principal Structural Materials Identified Other Structural Materials Probable	Principal Materials Issues Identified Other Key Issues?
Prismatic Gas-Cooled Reactors	500–850	TRISO Fuel Graphite Moderator High Cr and Martensitic Steel Structures Advanced High Temp Alloys	Mechanical behavior of SiC at high burnup Diffusion of fission products through SiC Creep, Swelling & Embrittlement of core internals Thermal or Radiation-Induced Toughness/DBTT
Pebble Bed Gas-Cooled Reactors	500–900	TRISO Fuel Graphite Moderator High Cr and Martensitic Steel Structures Advanced High Temp Alloys	Mechanical behavior of SiC at high burnup Diffusion of fission products through SiC Creep, Swelling & Embrittlement of core internals Thermal or Radiation-Induced Toughness/DBTT
Gas-Cooled Fast Reactors	500–900	TRISO Fuel Fuel Matrix (Unspecified) Structural Steel (Unspecified) Advanced High Temp Alloys	Identify fuel matrix material, behavior in fast spectrum Diffusion of fission products through SiC Creep, Swelling & Embrittlement of core internals Thermal or Radiation-Induced Toughness/DBTT
High Temperature Gas-Cooled Reactors	900–1250	Modified TRISO Fuel (ZrC) Ceramic Composite Structures Refractory, Intermetallic Alloy Structures Advanced High Temp Alloys High-Temperature Coatings	Thermal and dimensional stability Fabrication/reliability of ceramic composites Irradiation/ Mechanical behavior of ZrC Irradiation, creep, embrittlement for HT alloys
Molten Salt Cooled Reactors	1200–1600	TRISO Fuel Graphite Structures Advanced High Temp Alloys Ceramic Composites, High-Temp Coatings	Behavior of TRISO fuel at high temperature Environmental effects in fluoride salts Irradiation, creep, embrittlement for HT alloys Irradiation, reliability of ceramic composites
Gas Fueled Reactors	1500–2200	Refractory Carbide Fuel Matrix/Structure/Vessel Be/BeO Reflector Graphite Structures Refractory Alloy Structures Ceramics/Ceramic Composite Structures Ni-Al, Ni-Mg Alloy Vessel; Au, CaF_2 Vessel High-Temp, Corrosion-Resistant Coating	Synthesis of suitable refractory carbides Fabrication of structural ceramic composites Irradiation behavior of ceramics/composites Protective coatings for metallic vessels Environmental effects in UF_4/UF_6 Irradiation, creep, embrittlement for HT alloys Irradiation, reliability of ceramic composites
Plasma Fueled Reactors	>4000	Unspecified	Materials issues similar to tokamak first wall Thermal stability, thermal shock Environmental effects, erosion Many more issues !

- Corrosion in molten salts
- Compatibility of fuels and cladding

Additionally, several fundamental science issues were identified:

- The co-evolution of all components of the microstructure, and their roles in the macroscopic response in terms of swelling, anisotropic growth, irradiation creep, and radiation-induced phase transformations should be studied within the of the science of complex systems.
- Displacement damage during irradiation creates a non-equilibrium, structure-chemistry evolution at the nanoscale and alters plasticity, corrosion-oxidation and fracture processes. Fundamental understanding of these complex, interdependent, radiation-induced material changes is essential to underpin the development of Generation IV reactor systems.
- Key structural performance issues for most irradiated metallic alloys are time-independent embrittlement at low temperatures and time-dependent cracking at high temperatures. The evolution of non-equilibrium structures and chemistries promote a hardened matrix and lower grain boundary cohesive strengths thereby reducing the tensile stress required for cleavage or intergranular fracture. Advances in modeling and measuring the atomistics of fracture need to be combined with micromechanical models to better elucidate behavior in complex radiation-induced, multi-component nanostructures.

Readiness of Research Facilities

Proper study and evaluation of materials for Generation IV systems requires irradiation facilities and the facilities to prepare and analyze samples. Currently, the U.S. does not have a fast spectrum irradiation capability. Many Generation IV concepts that optimize recycle are based on fast spectrum systems. To irradiate materials in a fast spectrum requires the use of either the JOYO reactor in Japan, BOR-60 in Russia, or PHENIX in France. Thermal spectrum facilities exist in the U. S. at the Advanced Test Reactor at the INEEL and the High Flux Isotope Reactor at ORNL.

While neutron irradiations are essential to evaluate and qualify materials for Generation IV systems, it is important to note that effective radiation effects experiments can be performed using ion-beam facilities. Currently DOE NE is sponsoring work using ion-beam facilities at both the University of Michigan and PNNL. These facilities are good for studying microstructural and microchemical changes during irradiation as well as corrosion and mechanical properties in many circumstances. However, the higher dose rates must be taken into account and the depth of penetration is typically not sufficient to assess bulk mechanical properties. Charged particle irradiations can provide a low-cost method for conducting valuable radiation effects research in the absence of, or as a precursor to verification experiments in reactors. Some specialized facilities such as the ORNL triple beam facility and the ANL 1MeV electron microscope have been closed in recent years. As a result, existing facilities, while of high quality, are inadequate to handle the entire workscope required for a major alloy design and verification effort.

Dedicated hot-cell facilities are required to examine radioactive materials. Due to lack of support, many hot-cell facilities are being decommissioned. While not severely limiting materials R&D yet, without adequate programmatic support, more important hot-cell capacity may be lost in the near future. In addition to maintaining existing capabilities, it is important to note that most hot-cell facilities are more than 30 years old. Modern facilities for the handling and testing of radioactive materials are needed. A vital capability in this regard has been the development of small-specimen testing methods capable of generating high-quality data for bulk properties.

Readiness of Research Personnel

An increase in research on materials for advanced reactor concepts will require not only financial resources, but the human resources to carry out the plan. The latter may in fact become limiting in light of the shrinkage and aging of the workforce in the nuclear field over the past two decades. In the national laboratory community, only three labs (ORNL, ANL and PNNL) have a critical mass of scientists that can contribute in a significant way to research in materials for advanced reactor concepts. In academia, there are only about 15 faculty currently active in nuclear materials research in the nation's nuclear engineering departments, where most of this type of research is conducted. In industry, there are only a few companies with laboratory capabilities and manpower capable of conducting research on materials for advanced reactor concepts. If the goal is to develop a knowledge base of materials issues on more than once advanced reactor concept, the human resources in the US will be extremely strained.

Academia

In January of 2000, the Nuclear Engineering Department Heads Organization (NEDHO), published a report entitled "Manpower Supply and Demand in the Nuclear Industry" that contained a survey and assessment of manpower in the nuclear industry.[5] The Manpower Survey resulted in a quantitative assessment of both the supply and the demand for nuclear engineers out to the year 2003. The results show that the demand—supply gap increases monotonically from 1999 to 2003. The results of the survey are summarized below:

For the supply side:

- 29 nuclear engineering departments continue to award either the B.S. or the M.S. in nuclear engineering or both,
- Nuclear engineering departments report a relatively stable supply of NE graduates through the year 2000–2001,
- Nuclear engineering departments will collectively graduate an average of 110 B.S. students per year that have focused their studies in fission engineering, of which about 83 are expected to be available for employment in the nuclear industry,
- Nuclear engineering departments will graduate an average of 106 M.S. students per year that have focused their studies in fission engineering, of which about 80 are expected to be available for employment in the nuclear industry.

For the demand side, the survey results are summarized as follows:

1. 52% of organizations contacted responded to the survey
2. 91% of respondents expect to hire nuclear engineers within the next 5 years
3. 78% of organizations will hire fresh graduates
4. 74% will hire non-nuclear engineers with nuclear engineering knowledge

5. 61% are having difficulty recruiting nuclear engineers

6. The average yearly demand of 52% of the organizations contacted is 337 engineers with either a B.S. or a M.S. degree

7. Of 497 vacancies in 1998, 316 were filled by nuclear engineers.

The survey primarily assessed the production of and demand for BS and MS graduates in the fission reactor area. Much of the research on materials for advanced reactors will be conducted by PhDs in academia, national laboratories and nuclear-oriented companies, and needs to be included in any assessment of available human resources. To this end, the enrollment and degree production of all levels of nuclear engineers is shown in Figure 5. As noted, there has been a startling decrease in BS enrollment starting in the early 90s and leveling off at the end of the decade. The decrease is tempered somewhat at the MS level and more so at the PhD level. Nevertheless, the total amount of graduates represents a fairly low production rate that is insufficient to maintain the current level of research activity across the various organizations, let alone accommodate an expansion of research.

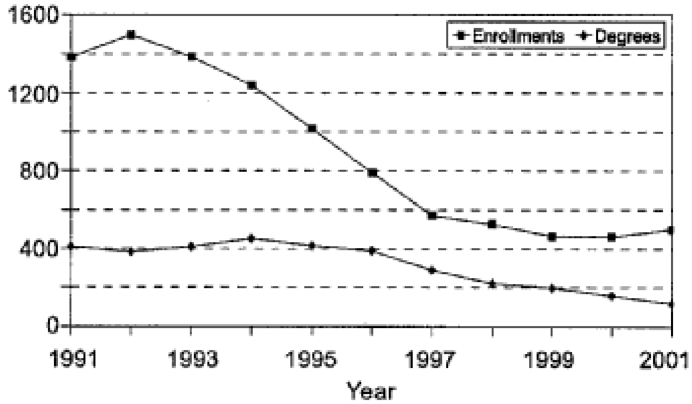
Not all of the manpower for advanced reactor materials will come from the nuclear engineering discipline. Yet there are similar concerns regarding the availability of talent to address an increase in research. Figure 6 shows undergraduate degree production and graduate enrollment in several fields including life sciences, physical sciences and engineering in the U.S. over the past 10-15 years. Over this time period, there has been tremendous growth in the graduate student population in the life/biological sciences and a decline in population of engineering and the physical sciences. Figure 7 shows that the graduate program enrollment in science and engineering is further challenged by a declining domestic student population, and increasingly buttressed by foreign graduate students. Since Generation IV Systems critically rely on advances in materials, energizing an interest among material science departments is an important step.

Industry

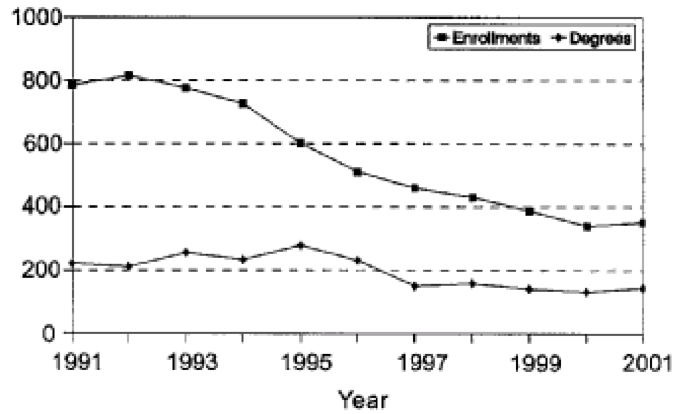
In 2001, the Nuclear Energy Institute (NEI) completed an extensive survey focused on the staffing and recruiting projections from 2002 to 2011, called the Nuclear Industry Staffing Pipeline Survey.[6] In consultation with a diverse group of individuals representing various sectors, they identified actions underway or planned that support building a reliable workforce pool of entry-level workers for the industry over the next decade and beyond. This document was intended to be fluid and flexible—allowing growth in concepts and programs as more is known and progress occurs. Key conclusions from the survey included:

- The industry’s demand for new staff will increase dramatically over the next 10 years. Approximately 90,000 entry-level workers are estimated to be needed to fill vacancies in thirteen job classifications simply to support the existing operations. Demand for graduates in nuclear engineering will be 50–100% greater than the supply.

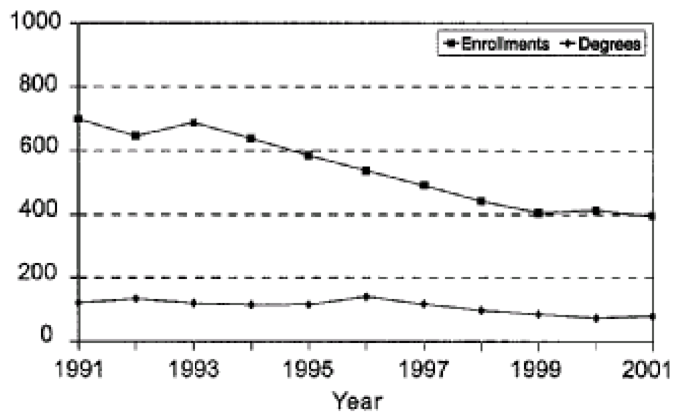
- The demand for degreed health physicists and nuclear engineers, although small by comparison in numbers, will significantly outstrip the supply for the next decade. A particular difficulty in employing degreed health physicists is that the demand for these candidates extends well beyond the nuclear energy industry. The shortage in nuclear engineers is expected to equal about 800 over the next 10 years.



(a)

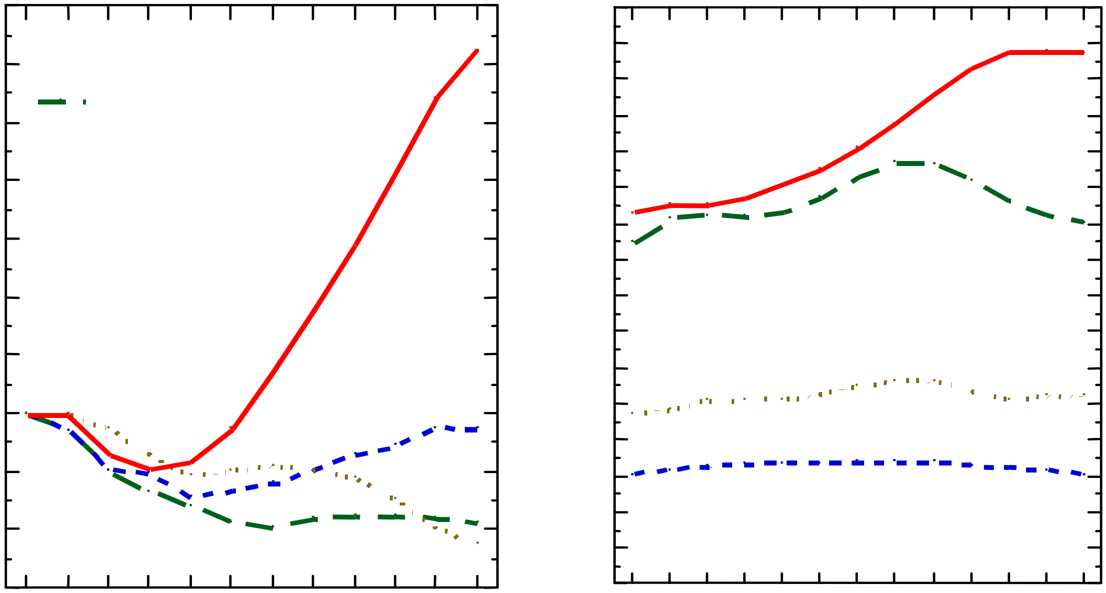


(b)



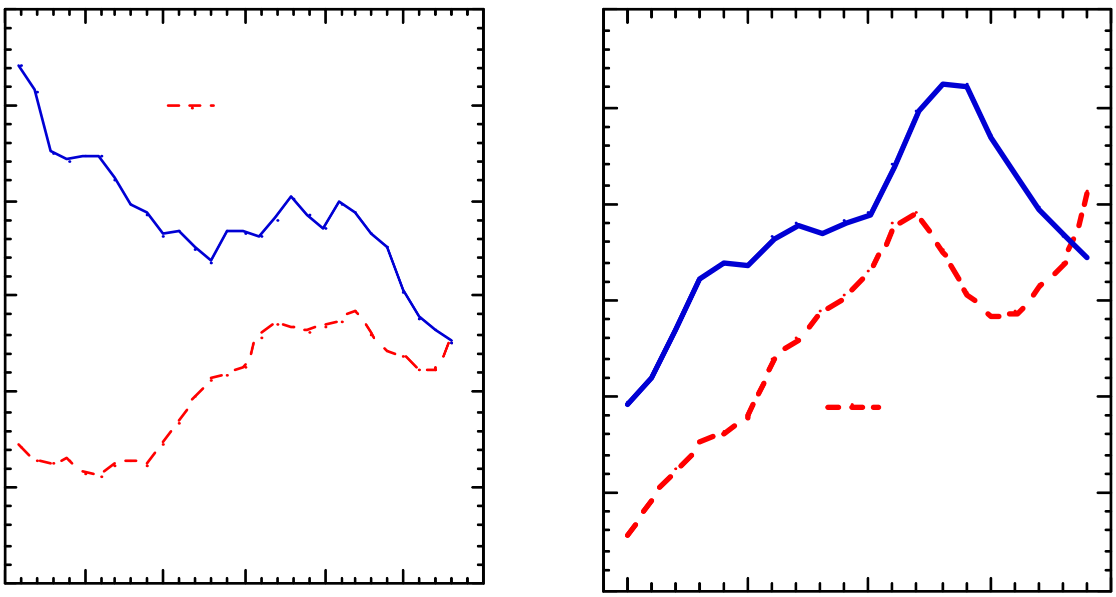
(c)

Figure 5. Degree production and enrollment in (a) BS, (b) MS and (c) PhD programs in nuclear engineering in the U.S. through 2001.[8]



(a) (b)

Figure 6. Increases in (a) undergraduate degrees and in (b) graduate enrollment in the biological/life sciences, physical sciences, mathematics and engineering.



(a) (b)

Figure 7. Change in numbers of foreign and U.S. graduate students in (a) physics and astronomy and (b) engineering between 1970 and 2000.

- While the availability of other degreed professionals, operators and skilled craft is adequate, stiff competition with other industries is expected. And, the experience and incoming qualifications of these entry-level workers are likely to be less.

- The nuclear energy industry is in a good position to compete for qualified employees in the current and future job markets. The industry is competitive in terms of overall employee compensation and in the six benefit categories that were examined in the study.

National Laboratories

Detailed reports for national laboratory manpower were not located in the process of writing this report. A recent report for DOE from the Laboratory Directors[7] did note that, even after accounting for differences in degree levels, the DOE Labs have a significantly smaller proportion of scientists and engineers under the age of forty than the US norm (26% vs 40%). Additionally, the national laboratories had very high attrition rates (greater than 8%) for scientists 34 years old and younger. Anecdotal evidence indicates that DOE laboratories will have staffing shortages similar to academia and industry in meeting the demands of a significant growth in R&D to support Generation IV.

Reactor Materials Areas not Specifically Addressed

To control the workshop scope to a manageable level, the workshop did not address all of the issues that significantly weigh on the choice of materials for Generation IV systems. Specifically, the following items relative to structural materials were not addressed:

- Chemical compatibility and corrosion issues

- Material availability, cost, fabricability, joining technology

- Safety and waste disposal aspects (decay heat, etc.)

- Nuclear properties (neutron economy, solute burnup, etc.)

All of these issues play a critical role in determining if reactor concepts can be operated safely and reliably and at a reasonable cost. Because many of the proposed Generation IV concepts operate in unique coolants (e.g. lead, lead-bismuth, supercritical water, molten salt), research and development will be required to establish materials that can operate in these environments. Material availability and cost is certainly an issue. Table 6 demonstrates the varied costs associated with possible Generation IV materials.

Another major technology area not discussed was reactor fuels and fuel-cladding compatibility. Fuels are critical to Generation IV concepts, especially considering that sustainability is considered an important Generation IV goal. Fuels that are compatible with recycle systems and stable waste forms must be developed.

Materials issues in recycle systems were recognized as important, but were not discussed in depth at this workshop. To be economically viable, recycle systems must minimize process losses. Materials improvements in all steps of the recycle process are needed.

Table 6. Costs for simple plate products (1996 prices).[2]

Material	Cost per kg
Fe-9Cr steels	≤$5.50 (plate form)
SiC/SiC composites	>$1000 (CVI processing)
	~$200 (CVR processing of CFCs)
V-4Cr-4Ti	$200 (plate form--average between 1994-1996 US fusion program large heats and Wah Chang 1993 “large volume” cost estimate)
CuCrZr, CuNiBe	~$10
Nb-1Zr	~$100
Ta, Ta-10W	$300 (sheet form)
Mo	~$80 (3 mm sheet); ~$100 for TZM
W	~$200 (2.3 mm sheet); higher cost for thin sheet

Recommendations for Future Workshops

This workshop proved to be a useful first discussion about the materials aspects of Generation IV nuclear energy systems. Because Generation IV reactor systems were not well enough defined at the time of the workshop to identify specific operating environment, the workshop took a rather broad view of materials issues. At the completion of the Generation IV Roadmap, the number of concepts being considered will be fewer than those considered at this workshop. Future workshops should aim to discuss a narrower issue in greater technical depth.

The workshop participants noted that in the area of reactor structural materials for Generation IV systems, many of the alloy systems to be considered are also being considered in the fusion reactor development community. Coordination of future workshops and R&D plans with the fusion reactor community would prove useful and optimize R&D resources.

Possible topics for future workshops include:

- Coolant specific corrosion and environmental cracking issues relative to specific Generation IV concepts (e.g., in lead-base, molten-salt, or supercritical water coolants)
- Materials to minimize process loss in fuel recycle systems
- Fuel development for specific Generation IV systems (e.g., nitride fuel development)

Because R&D on specific Generation IV concepts will start in the fall of 2002 at the earliest, a workshop in the Summer/Fall of 2003 is likely to be properly timed to ensure significant issues and results can be discussed.

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